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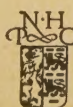
NUCLEAR INSTRUMENTS & METHODS

A JOURNAL ON ACCELERATORS, INSTRUMENTATION
AND TECHNIQUES IN NUCLEAR PHYSICS

EDITOR: K. SIEGBAHN - UPPSALA

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A journal on accelerators, instrumentation and techniques in nuclear physics

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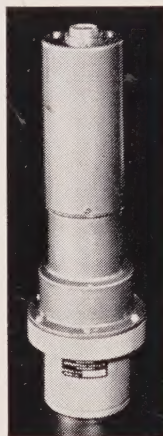
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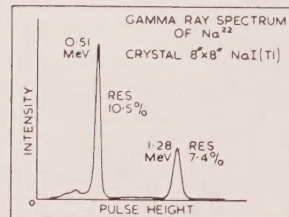
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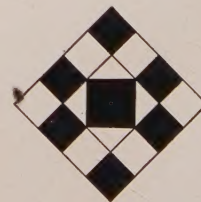
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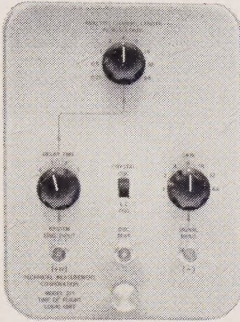
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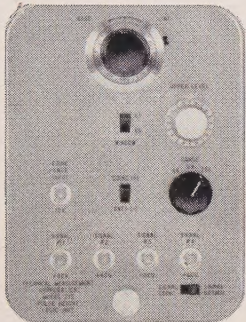




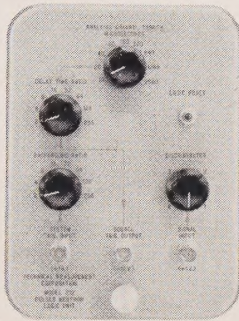
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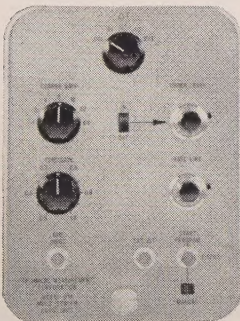
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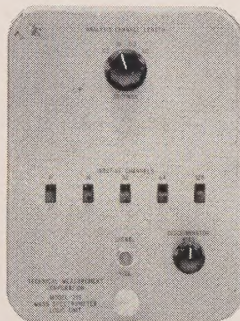
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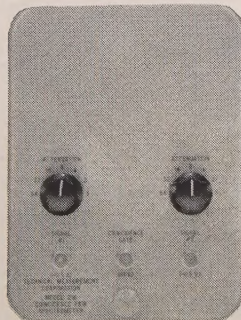


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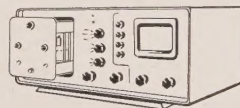
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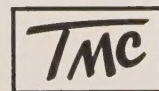
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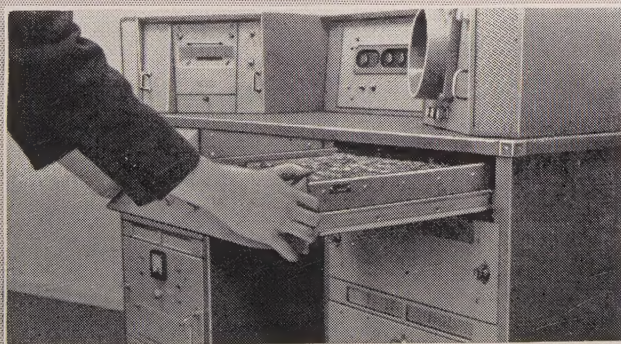
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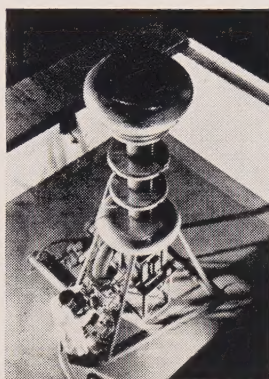
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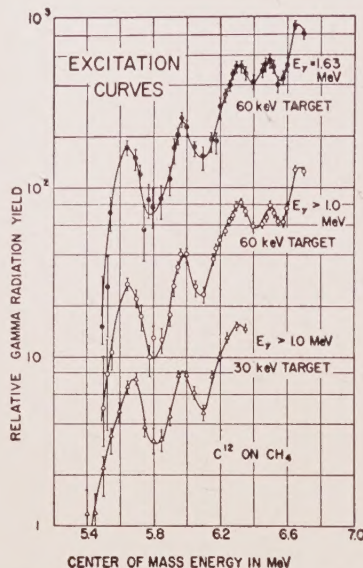
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A paper at our recent Accelerator Conference, “Current Experimentation with the Tandem Accelerator at the Chalk River

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“Low-Energy” Physics

As we address ourselves to this subject, more elegantly called *nuclear-structure physics*, the reader may conclude we have an axe to grind, and we admit it. We believe a great deal of research remains to be done on light nuclei. There is, for example, time-consuming but rewarding precision nuclear spectroscopy to fill in gaps in existing energy level data, as well as new research related to the conservation of isotopic spin, excitation energies of low excited states and direct interaction mechanisms.

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¹H. E. Gove, *Proceedings of the Second Accelerator Conference, Amsterdam, Oct., 1960* (North-Holland Publishing Company, 1961) p. 63.

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COLLECTION AND SPUTTERING EXPERIMENTS WITH NOBLE GAS IONS

O. ALMÉN and G. BRUCE

Department of Physics, Chalmers University of Technology, Gothenburg

Received 9 January 1961

The present paper describes experimental work on the collection of noble gas ions of energies 5–65 keV. We have used an electromagnetic isotope separator to measure saturation values in different materials. The dependence of saturation value on different experimental conditions is investigated, e.g. variations with current density, angle of ion beam incidence and target

temperature. In order to provide a simple description of the collection process, we have also measured the sputtering ratios for noble gas ions bombarding different materials. Sputtering is discussed in the light of existing theories for which a few improvements are proposed.

1. Introduction

In electromagnetic isotope separators of medium size¹⁾ the isotopes are usually collected by the direct collection method. This means that the ion beam hits a metal plate and some of the penetrating ions are trapped in the metal lattice. This collection technique has been used by several authors to produce targets for beta-gamma spectroscopy and charged particle bombardments.

For the collection of noble gas isotopes^{2,3,4)}, the direct collection method is the only possible way to collect ions. A few measurements of collected quantities are described in literature. E.g. the collection of argon in some backing materials has been studied by Greene⁵⁾. The use of tracer technique in collection studies was first described by Thulin⁴⁾, who made a few collection experiments in connection with the preparation of Kr and Xe targets for beta-gamma spectroscopy.

In our laboratory it has been of great importance to find backing materials with a high saturation value for a work on (p, γ) reaction studies in neon and argon^{6,7)}. Therefore we have investigated in detail the collection of different noble gas ions in many backing materials. In order to reach a deeper understanding of the collection process, we have also measured the sputtering ratio of noble gas ions bombarding different materials.

Sputtering phenomena have been studied by many authors. Much of the early work has not been made under reliable experimental conditions, but recent authors^{8,9,10)} have managed to bring under control

most of the parameters involved. We have used the electromagnetic isotope separator at our laboratory¹¹⁾ for all our collection and sputtering experiments. In this way we obtained the following experimental conditions:

1. ions of definite energy in the range 5–50 keV and sometimes up to 65 keV;
2. ions of definite mass and charge;
3. ions of a definite entrance angle, which can be easily varied;
4. a low pressure in the target chamber, giving no back diffusion of sputtered particles.

All these conditions are necessary to get reliable results in sputtering experiments. Therefore one must be cautious, when comparing results obtained by different authors.

2. Qualitative Description of Direct Collection of Noble Gas Ions

At direct collection the ion beam strikes a cleaned surface of the backing material, usually

¹⁾ J. Koch, Proc. 1955 Harwell Isotope Conf., Ch. 25.

²⁾ K. J. Broström, T. Huus and J. Koch, Nature **160** (1947) 498; and J. Koch, *ibid.* **161** (1948) 566.

³⁾ I. Bergström, Arkiv Fysik **5** (1952) 191.

⁴⁾ S. Thulin, Arkiv Fysik **9** (1954) 107.

⁵⁾ H. B. Greene, ORNL 2275.

⁶⁾ S. E. Arnell, J. Dubois and O. Almén, Nucl. Phys. **6** (1958) 196.

⁷⁾ S. E. Arnell, Nucl. Phys. **24** (1961) 500.

⁸⁾ F. Keywell, Phys. Rev. **97** (1955) 1611.

⁹⁾ G. K. Wehner, Phys. Rev. **102** (1956) 690.

¹⁰⁾ P. K. Rol, Thesis (Amsterdam, 1960).

¹¹⁾ O. Almén, G. Bruce and A. Lundén Nucl. Instr. **2** (1958) 249.

perpendicularly. Every impinging ion undergoes a slowing down by a series of collisions and can either be backscattered, after one or more collisions, or be trapped in the metal lattice. The mean depth, $\bar{d}$, at which an ion is trapped is determined by the ion energy and the mass and atomic numbers of the ion and the target atoms (Nielsen¹²). The probability for an impinging ion to be trapped is η_i and we assume η_i to be independent of the amount of ions already collected. At the beginning of the collection, η_i is identical with the collection efficiency. When the ratio f between the number of collected ions and the total number of particles per unit volume is increased, previously trapped particles can take part in the collision process of an impinging ion. Therefore trapped atoms can diffuse out or be released together with sputtered target atoms. After sufficiently long bombardment time, saturation of the target is reached and this means that for every collected ion one gas atom must leave the target, or in other words, for every impinging ion η_i atoms must leave.

We make the following assumptions:

At saturation, D is the fraction of impinging ions, which leaves due to diffusion. Then $\eta_i - D$ is the fraction leaving because of sputtering. Since the sputtered material must have the same composition as the outer layer of the target, f_s , we can write

$$f_s = \frac{\eta_i - D}{\eta_i - D + S} \quad (1)$$

where S is the number of atoms of the original target material sputtered off for every impinging ion. S is called the sputtering ratio.

If we measure the amount of material collected at saturation, $q \mu\text{g}/\text{cm}^2$, and use the penetration depth $\bar{d} \mu\text{g}/\text{cm}^2$ given by Nielsen¹²) we can approximately calculate for the penetration depth, the mean ratio $\bar{f}$ between the trapped ions and all particles. We get

$$\bar{f} \approx \frac{q/M_1}{q/M_1 + \bar{d}/M_2} \quad (2)$$

where $\bar{d}$ is used as an approximation of the real penetration depth, which is affected by the trapped atoms. M_1 , M_2 are mass numbers of ion and target

atoms, respectively. f_s must, however, be lower than $\bar{f}$, since the concentration of collected particles falls off towards the surface owing to diffusion. With a proper assumption of the relation between f_s and $\bar{f}$, together with measured values of S and η_i we can estimate D , see § 11.

3. Determination of Saturation Values

3.1. Kr AND Xe IONS

The two noble gases Kr and Xe have radioactive isotopes of half-lives long enough to be suitable for collection experiments using the tracer method. Both Kr^{85} (10.6 year, 0.67 MeV beta and practically

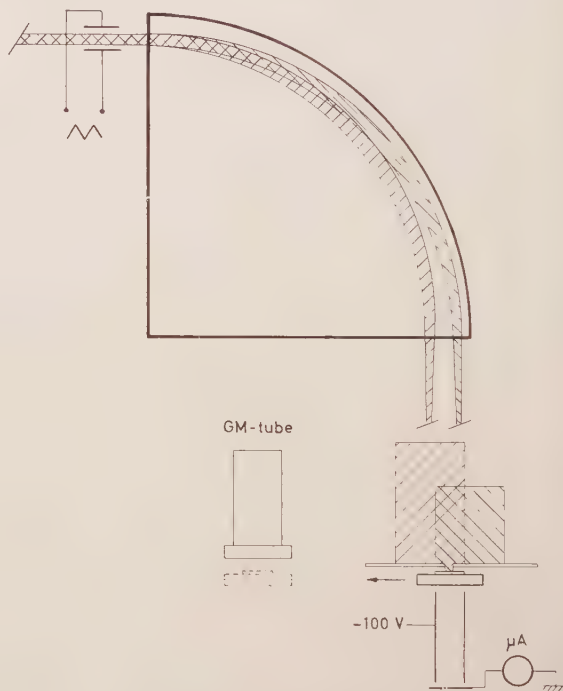


Fig. 1. Collector arrangement for saturation studies by means of radioactive tracers.

no gamma radiation) and Xe^{133} (5.27 d, 0.34 MeV beta and a very weak gamma radiation) are fission products available in quantities up to several curies. 100 mc of each isotope, delivered from Harwell, were mixed with about 0.5 l NTP inactive Kr and Xe gas, respectively, and fed to the ion source of the electromagnetic isotope separator.

By sweeping the analyzed beam over the collector, the radioactive isotope was mixed with an

¹²) K. O. Nielsen, Proc. 1955 Harwell Isotope Conf., Ch. 9.

adjacent isotope giving a current density of 2–15 $\mu\text{A}/\text{cm}^2$. This density is of a magnitude often used at the collection of stable isotopes in a separator of medium size. The method is shown schematically in fig. 1. The linear sweep is produced by means of two plates at the entrance side of the separator magnet, which are fed with a saw-tooth voltage of a few hundred volts. Since the target area, where collection was studied, was chosen much smaller than the total area of the modulated beam, a smooth current density was secured over the target.

The arrangement for observing the increase of collected quantity is also shown in fig. 1. In the position to the right the target is hit by the ion beam. After a few minutes of collection the target is moved to a measuring position, where the activity is determined by a GM-tube and a scaler. At the same time the current, entering the opening in the front cover, is measured in a Faraday cage. By alternatively collecting and measuring we can follow the increase of the noble gas quantity in the target material. Fig. 2 shows a typical collection curve, which will be discussed below. Four target plates were mounted on a slide and could be bombarded successively.

Outside the separator the activity of a target was measured in a standard geometry and compared with an Al target, saturated with the gas in question. The quantity collected in this reference target was determined in another way (see below). A correction for different backscattering of the electrons from different materials must be made before the collected quantity can be calculated. The correction factor was determined directly from the change of activity, when plates of different material were placed behind a 150 $\mu\text{g}/\text{cm}^2$ Al foil, containing radioactive Kr. The decrease in counting rate because of the passage through the Al foil was negligible.

Absolute quantities collected in some metal foils were determined in the following way:

First a small quantity of the radioactive gas mixture was isolated in a glass vessel of known volume (a few cm^3) and the gas pressure was measured (about 500 mm Hg). Then the gas was let out into a container with about 500 times larger volume. The

quantity remaining in the glass vessel was brought into a measuring container in which there was a thin window of a 0.05 mm Al foil. The activity of this gas was measured with a GM-tube outside the window.

After that the noble gas collected in a metal foil was released in the measuring container by thoroughly outgassing the foil. The new activity measured outside the window gave the collected quantity in $\mu\text{g}/\text{cm}^2$. We observed that the foil had to be heated up to just below the melting point to release all enclosed gas. The rest activity was measured in all cases and the result was corrected, if there was a measurable rest. All measurements were corrected for drift of the GM-tube sensitivity and of the scaler by using a reference activity.

Mean values of several absolute determinations of saturation values are given for different materials in table 1, using the two noble gases Kr and Xe. From the results in table 1 and the relative measurements the gas quantity collected in an Al foil was calculated and its activity was used as a reference target, as mentioned above.

TABLE 1
Saturation values of Kr and Xe at 45 keV ion energy in some metal foils.

Target material	Kr ($\mu\text{g}/\text{cm}^2$)	Xe ($\mu\text{g}/\text{cm}^2$)
Ti	4.7	3.3
Ni	2.2	1.9
Co	2.3	1.5
Mo	3.2	2.0
Ta	3.1	—
Pt	1.4	1.3
Al reference	5.2 $\mu\text{g}/\text{cm}^2 \pm$ 5 per cent	4.1 $\mu\text{g}/\text{cm}^2 \pm$ 5 per cent

Saturation values may also be calculated directly from the collection curves. The method is indicated in fig. 2. It was noted that collection curves for many materials have approximately the same slope at $t = 0$, proportional to current density. If the target mass M_2 is less than ion mass M_1 , all ions are expected to penetrate the target surface and the collection efficiency at $t = 0$, η_i , should be equal to 1. If M_2 is larger than M_1 head-on collisions on surface atoms should decrease η_i . In

§§ 3.2 and 6 we shall see that η_i can be nearly 1 even when $M_2 > M_1$.

If we suppose that we reach the saturation value along the straight line through origin, which represents the efficiency η_i , (see fig. 2), the saturation value q can be calculated from the current density I (in $\mu\text{A}/\text{cm}^2$) and the time t_1 (in hours). We get

$$q = 0.0376 \eta_i I M_1 t_1 \quad \mu\text{g}/\text{cm}^2. \quad (3)$$

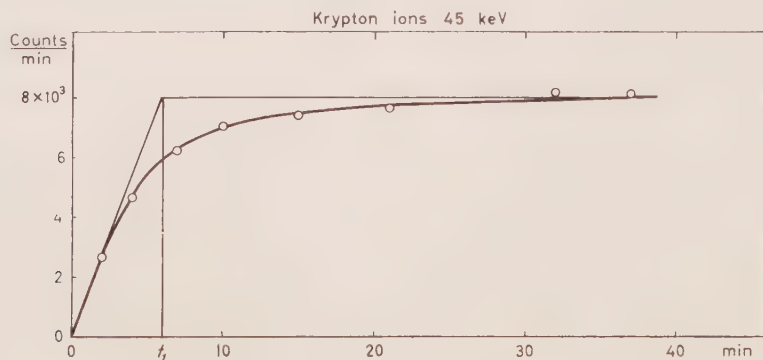


Fig. 2. Typical collection curve for a low sputtering metal, Mo.

For combinations where η_i is known this method will give saturation values equal to those determined by the radioactive method. Experimental values of η_i are given in § 6.

3.2. Ne AND Ar IONS

In the collection studies of Ne and Ar ions a different technique from that used for Kr and Xe must be applied. If a Ne^{20} or an Ar^{40} target is

bombarded with protons the following reactions take place:



When the proton energy is varied several resonances appear in the intensity of the gamma radiation which is detected by means of a NaI-crystal and a photomultiplier⁷). One of the resonances from Ne^{20} is shown in fig. 3. It can be easily shown that the area under the resonance

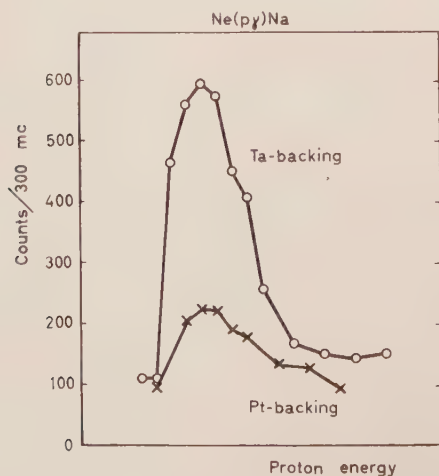


Fig. 3. 1.165 MeV (p,γ) resonance from Ne^{20} .

curve is proportional to the number of Ne^{20} atoms present in the target material, independent of the energy spread of the high voltage machine and also independent of the resonance width. If a resonance is measured on several targets of the same material, but bombarded in the isotope separator with varying amounts of $\mu\text{A}/\text{cm}^2$, and the areas under the resonance curves are plotted in a diagram, we get a collection curve similar to that shown in fig. 2 (see fig. 4). From this collection curve a saturation value can be calculated according to the method

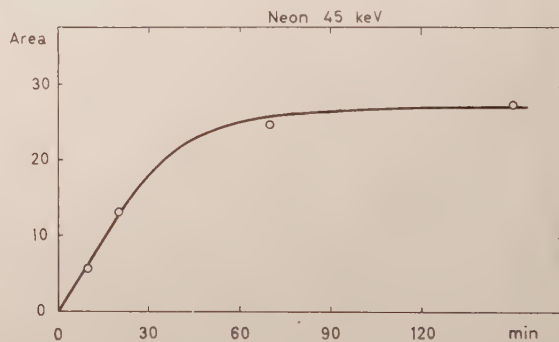


Fig. 4. Collection curve for Ne ions determined by means of (p,γ) resonance measurements.

discussed above, provided that $\eta_i \approx 1$ also for Ne and Ar. Thus in a Ta target the saturation values for Ne are: $q \approx 5.5 \mu\text{g}/\text{cm}^2$ and for Ar: $q \approx 3.5 \mu\text{g}/\text{cm}^2$ at 45 keV ion energy and $10 \mu\text{A}/\text{cm}^2$.

These two determinations in Ta can be used to get saturation values in other materials. For this purpose a (p, γ) resonance was measured on targets of different materials saturated with Ne or Ar, respectively. The Ne and Ar quantities collected in Ta were used as references. The areas under the

mined in Mo and the relative measurements for some other backing materials we calculated the saturation values given in table 2.

The reason why Mo was used instead of Ta for the mass spectrometer measurements was the difficulty to heat Ta to a temperature high enough for complete outgassing. As seen in table 2, the measured q in Ta is about the same as the value calculated from fig. 4. This means that η_i is close to 1 also for Ne and Ar collection in Ta.

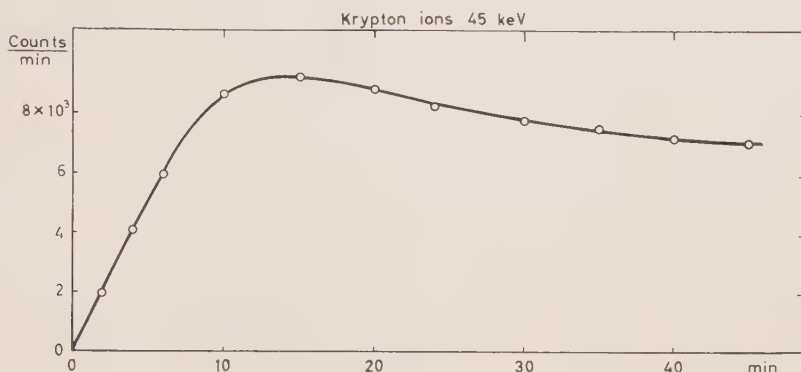


Fig. 5. Collection curve for Mg backing.

resonance curves (background subtracted) are proportional to the collected quantities.

We have also used a mass spectrometer to get saturation values of Ne and Ar. In this case a Mo foil was bombarded with Ne ions on one side and with Kr ions on the other. After saturation was obtained with the two gases the foil was outgassed in an evacuated container. The released gas was fed to the ion source of a mass spectrometer and the ratio between the two ion beams was measured. Since the two gases have different cross sections for ionization it was necessary to measure the ion beams, when a gas mixture containing known quantities of the two gases was fed to the ion source. These two measurements gave the ratio between saturation values of Ne to Kr and Ar to Kr. In addition, we determined the Kr^{86} quantity collected in a Mo foil by measuring the change of isotopic abundance, when natural Kr was mixed with Kr^{86} released from a Mo foil. The result is in agreement with the saturation value determined by means of radioactive Kr.

From the Ne and Ar absolute quantities deter-

TABLE 2

Saturation values of Ne and Ar at 45 keV ion energy in some metal foils. For comparison the mass spectrometrically determined value of Kr collected in Mo is also given.

Target material	Kr ($\mu\text{g}/\text{cm}^2$)	Ne ($\mu\text{g}/\text{cm}^2$)	Ar ($\mu\text{g}/\text{cm}^2$)
Ni	3.0	2.1	2.4
Cu		1.7	1.2
Mo		4.7	3.8
Ag		0.8	0.8
Ta		5.0	3.7
W		4.5	2.5
Pt		1.7	1.6
Au		1.0	0.7

3.3. ACCURACY AND REPRODUCIBILITY

In all target materials small variations of the saturation value were found depending on the inhomogeneous structure of the material and also on the varying thickness of the oxide layer. E.g. a difference is sometimes observed between rolled material and material annealed in vacuum after rolling. Normally all these variations are within

± 10 per cent. Our relative measurements with Kr and Xe are reproducible within about ± 3 per cent, if the same target material is used, but with respect to the variations mentioned, most results given in § 4 must be regarded as the mean of several

electrolytic deposition or by vacuum deposition. E.g. the saturation value was lower by a factor of two in electrolyzed than in rolled Ag.

In fig. 5 a collection curve is shown for a Mg backing material. The collected amount increases

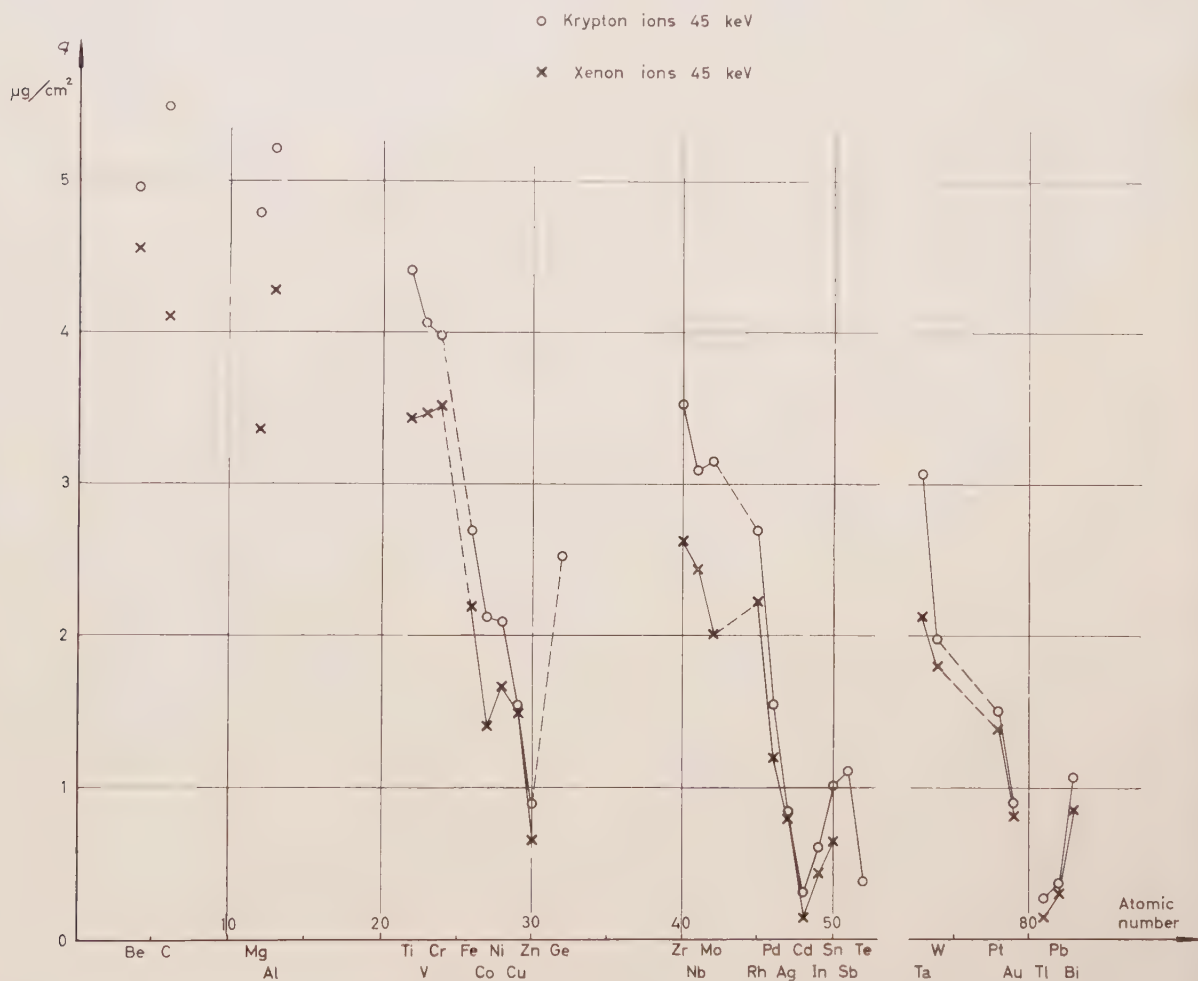


Fig. 6. Saturation values of Kr and Xe in different backing materials.

determinations for different qualities of a material. In addition we have an experimental error in the determination of the quantity collected in the reference foils, amounting to ± 5 per cent. The results for Ne and Ar are expected to be less accurate regarding relative as well as absolute determinations. The probable total error is ± 15 per cent.

On the other hand we have observed greater variations than ± 10 per cent between saturation values for rolled foils and for layers produced by

to a maximum and after a while decreases to a lower value. This effect is probably due to sputtering away of an initial oxide layer on the surface. The saturation values for Be, Mg, Zn and Hf decrease in this way during a prolonged bombardment. On the other hand Al, Ta and also other materials covered with a permanent oxide layer do not show this decrease at any of the current densities we have used. E.g. Al was bombarded 10 times longer than the normal saturation time without any change in the saturation value.

4. Saturation Values in Different Materials

Fig. 6 shows saturation values determined for some 30 target materials using Kr and Xe ions of 45 keV energy and a current density of $10 \mu\text{A}/\text{cm}^2$. Dependence on current density is treated in § 5.

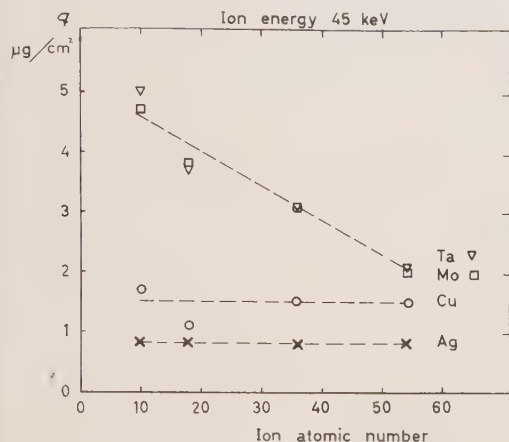


Fig. 7.

Saturation values for Ne, Ar, Kr and Xe collection.

Most of the materials used here were rolled foils of purest available quality. Ge, Sb, Te and Bi were used in the form of blocks. Be was vaporized in vacuum and used as a thin foil of about $100 \mu\text{g}/\text{cm}^2$. C was investigated using both blocks and vaporized foils, giving saturation values with Kr of 5.5 and $7.5 \mu\text{g}/\text{cm}^2$, respectively. Cr and Rh had to be electrolyzed, since foils were not available commercially.

In fig. 6 we see that the saturation value q varies regularly within the periods of the periodic table. In

the periods 4 (K–Br), 5 (Rb–J) and 6 (Cs–At) the experimental results permit a systematic study of q . One finds that q reaches maxima for elements belonging to the same group of the periodic table e.g. for Ti, Zr, Hf (group 4a) (q for Kr in Hf, $2.6 \mu\text{g}/\text{cm}^2$, not shown in fig. 6.) and probably for As, Sb, Bi (group 5b). On the other hand q is minimum for the group 2b elements Zn, Cd. (Hg not investigated). If we calculate $\bar{f}$ (the ratio between the number of trapped particles and all particles), we find that $\bar{f}$ varies approximately in the same way as the binding energy of the elements, which indicates a relationship between these two quantities.

In fig. 6 it is also to be noted that q for Kr is usually higher than q for Xe. In hard materials still higher q values are found for Ne and Ar (see fig. 7). If we assume that $\bar{f}$ varies slowly with the ion mass we see that this is in agreement with eq. (2) which can be written.

$$q \approx \bar{f} d \frac{M_1}{M_2}, \quad \text{since} \quad \frac{q}{M_1} \ll \frac{d}{M_2}.$$

In this equation a decrease in M_1 is more than compensated by the increase in d . (cf Nielsen¹².) However, in soft materials q is nearly independent of the ion mass. This is e.g. found for Cu and Ag (fig. 7) and also for Au. The explanation seems to be that for these materials there is a more pronounced dependence of ion mass on $\bar{f}$ caused by changes in diffusion (see eq. (1)). Of course there are target materials with ion mass dependences intermediate between the two cases above.

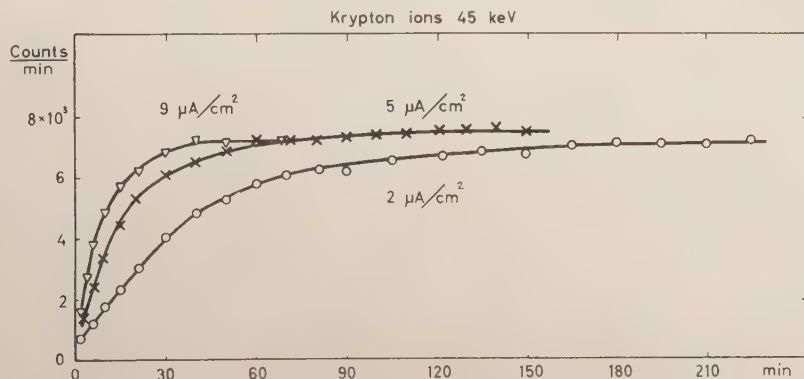


Fig. 8. Saturation curves for Al at different current densities.

5. Dependence of Operating Conditions on Saturation Values

5.1. CURRENT DENSITY

Fig. 8 shows how an amount of Kr is built up in an Al foil. It is to be observed that the collection curves are approximate exponential functions. We also notice that the maximum of the measured activity, thus the limit for collected Kr, seems to be

result would be obtained for Kr and Xe ions on Ta. This material is in fact one of the most suitable backing materials, as we shall see below.

However, in some cases current density determines the saturation value. In the experiment indicated in fig. 9 the current density was changed during operation by a factor of four. In an Ag foil a decrease in current density means a very small

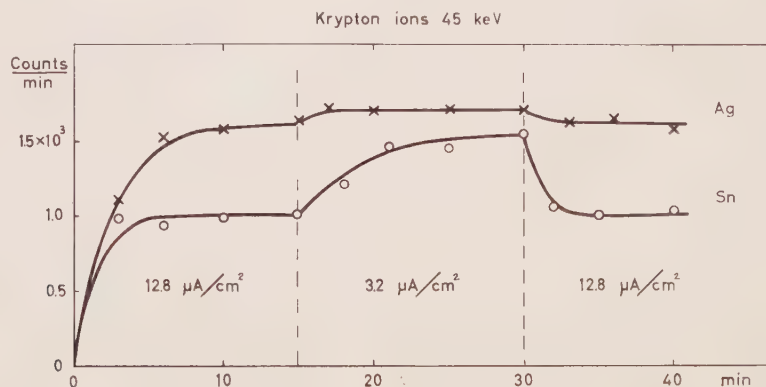


Fig. 9. Change of saturation value with current density.

independent of current density at least below $15 \mu\text{A}/\text{cm}^2$. Similar curves are obtained for Kr and Xe ions bombarding on most materials.

With Ar ions of 45 keV we have bombarded Ta to saturation using current densities of 10–150 $\mu\text{A}/\text{cm}^2$ and measured relative values by means of the (p, γ) resonance method. The targets were water-cooled during bombardment. The collected quantity was also in this case independent of the current density. It is to be expected that the same

increase of saturation value. In Sn, on the other hand, the increase is considerable. When the current density is increased again, the activity goes down to the original level. The observed variations are ascribed to diffusion of gas particles, which is determined by the surface temperature of the target.

5.2. TARGET TEMPERATURE

All measurements described so far were performed at a low target temperature 30–50°C, since cooling of the target was good and very little heat was dissipated in the foil, 0.1–1 W/cm².

In order to investigate the temperature influence on saturation values we made a series of collection studies on foils heated to different temperatures. Current density was the same as before, $10 \mu\text{A}/\text{cm}^2$. The foil was cooled to room temperature within about 5 minutes after the collection was finished, so there was no appreciable decrease because of diffusion after collection. Relative collection values are given in fig. 10, where the quantity collected at room temperature is chosen as a reference. As expected the collected quantity decreases when temperature is raised.

Our experimental equipment does not permit a

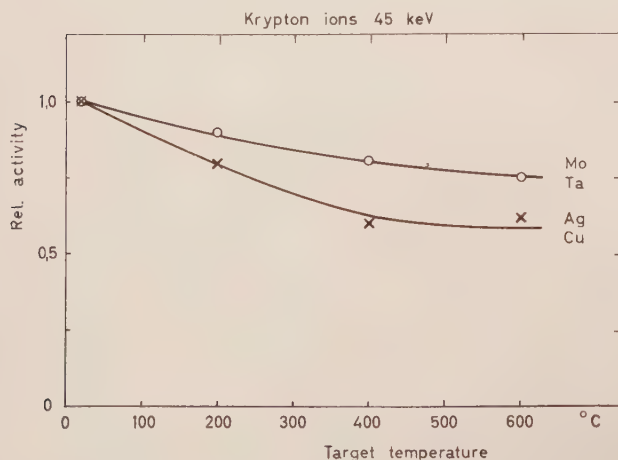


Fig. 10. Influence of target temperature on saturation values.

higher current density than $15 \mu\text{A}/\text{cm}^2$, when tracer technique is used. A high current density means a high surface temperature of the collector, much higher than that measured on the back of the collector. Therefore, our Kr measurements cannot predict the result, if the current density is much higher than this value. However, it is seen from

ion energy. If the number of gaseous particles which can be stored per unit volume in an atomic lattice is independent of the ion energy, the collected quan-

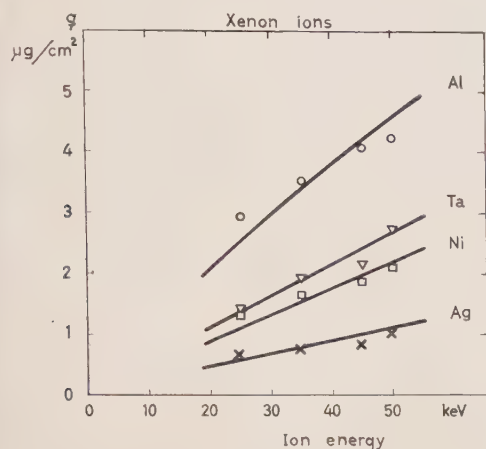


Fig. 11a.

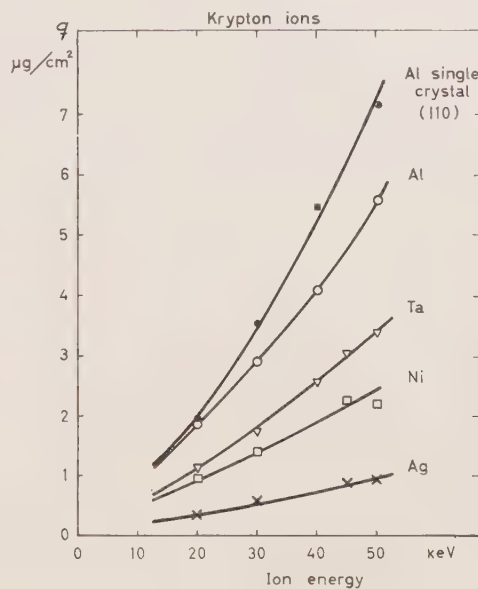


Fig. 11b.

Fig. 11 Saturation value versus ion energy.

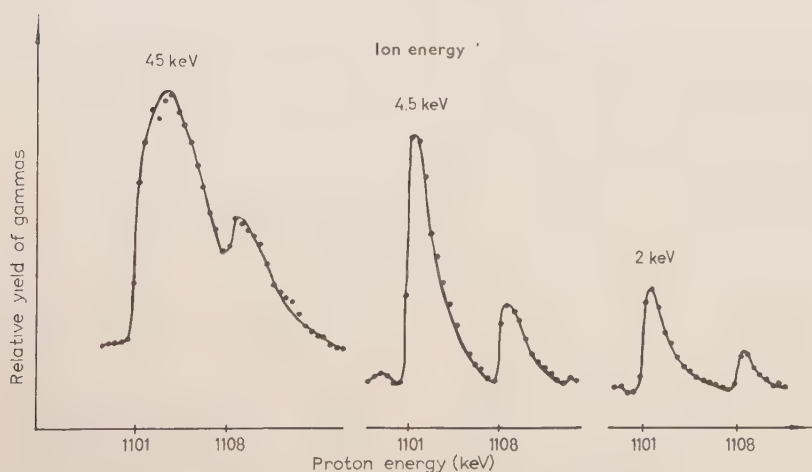
Fig. 12. 1.101 and 1.108 MeV (p, γ) resonances obtained from Ar targets prepared at different ion energy. (Cf ref.7.)

fig. 10 that it may be detrimental for a good collection of noble gas ions if the temperature is allowed to increase too much.

5.3. ION ENERGY

In the ion energy range used here, the penetration depth of atomic particles is proportional to the

tivity in a material should be proportional to the ion energy.

Figs. 11 a and b show that q increases approximately proportional to the energy. Deviations from linearity have been found to be due to bad focusing of the ion beam at low energies and also to a small non-linearity of the sweep voltage.

In fig. 12 (p, γ) resonance curves are measured on Ar targets prepared at three different energies of the Ar ions. We observe that the heights of the 45 and 4.5 keV curves are the same (background subtracted), while the 2 keV curve is much lower. This means that for ion energies between 45 and 4.5 keV the maximum concentration of Ar atoms in the backing material is about constant, which confirms the assumption above. At lower energies the penetration depth is so much reduced that collected particles can leave by diffusion.

We may also note the considerably increased resolution of the resonances, when ions are collected at low energy. To obtain this energy the impinging ions were retarded at the collector. The target was simply put at high tension behind a defining slit.

5.4. ANGLE OF ION BEAM INCIDENCE

In fig. 13 the influence on q of the angle φ between the ion beam and the normal to the target

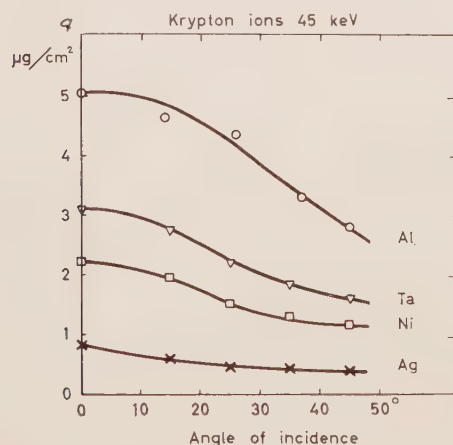


Fig. 13.

Saturation values at different angles of ion beam incidence.

plate is shown for some materials using Kr ions (see also fig. 23.) A first estimate gives that q is proportional to $\cos \varphi$, since the real penetration depth perpendicular to the surface should be approximately $d_0 \cos \varphi$. However, the saturation also depends on the sputtering ratio which increases with the angle of incidence (see § 8.6). Therefore we expect a faster decrease than a cosine law.

The best agreement with the experiments is given by

$$q_{\varphi} = q_0 \cos^2 \varphi.$$

This relation is most accurate for $M_1 > M_2$. If $M_1 < M_2$ and the entrance angle is large enough, the first collision can result in an increased velocity component of the ion perpendicular to target surface. This will result in a larger penetration depth and a higher saturation value than $q_0 \cos^2 \varphi$ for larger values of φ .

6. Collection Efficiency

The total efficiency of a collection is determined by the number of ions which have reached the collector, the saturation value and the initial efficiency η_i . If we assume that the saturation curve can be approximated by an exponential function (see § 5.1.), we can calculate the total efficiency for different degrees of saturation. Some values are given in table 3.

TABLE 3

Degree of saturation	0.40	0.63	0.78	0.87	0.95	0.98
Total efficiency	$0.80\eta_i$	$0.63\eta_i$	$0.52\eta_i$	$0.44\eta_i$	$0.32\eta_i$	$0.25\eta_i$

Normally there is no reason to collect to more than about 95 per cent of saturation. If η_i and q are known, we can easily get the necessary time of collection. For 95 per cent saturation we need 3 "time constants", which gives (cf eq. (3))

$$t = \frac{3q}{0.0376 \eta_i I M_1}. \quad (4)$$

TABLE 4

Initial collection efficiency for Kr ions of 45 keV. Experimental error in η_i is about 0.1.

Backing material	η_i	Backing material	η_i
Be	1.0	Mo	0.95
C	1.0	Rh	0.70
Mg	1.0	Pd	0.60
Al	1.0	Ag	0.35
Ti	1.0	Cd	0.20
V	1.0	In	0.70
Cr	1.0	Sn	0.50-0.70
Fe	0.95	Sb	0.80
Co	0.85	Ta	1.0
Ni	0.85	W	0.80
Cu	0.75	Pt	0.50
Zn	0.75	Au	0.35
Ge	0.80	Tl	0.30
Zr	1.0	Pb	0.30
Nb	1.0	Bi	0.50

In § 3.1 we have pointed out that the initial collection efficiency is in many cases equal to 1, also when $M_1 < M_2$. In table 4 we give η_i for Kr ions of 45 keV impinging on different materials, calculated from saturation curves and eq. (3). η_i for Xe ions is approximately the same. For Ne and Ar, measurements of η_i have been made only for Mo and Ta, giving about the same η_i as for Kr.

The periodicity of η_i indicates that a diffusion process of gas atoms rather than expected head-on collisions in the surface sets a limit for η_i . Since the mean free path of an ion after a head-on collision is strongly decreased, the ion has a very small chance to return directly.

7. Time and Temperature Stability of Noble Gas Targets

In collection of gaseous isotopes in a solid backing it is of great importance to know if the collected quantity decreases with time and what temperature it can stand during charged particle bombardment.

Fig. 14 illustrates an investigation, where Kr was collected in Al and Cu. After 30 minutes the ion beam was shut off and the measurement of the activity change was started, already in the collec-

ditional time of several days. After two hours, air was let into the collection chamber, and the comparing measurements were continued for another 10 days.

From fig. 14 it is seen that a Kr quantity col-

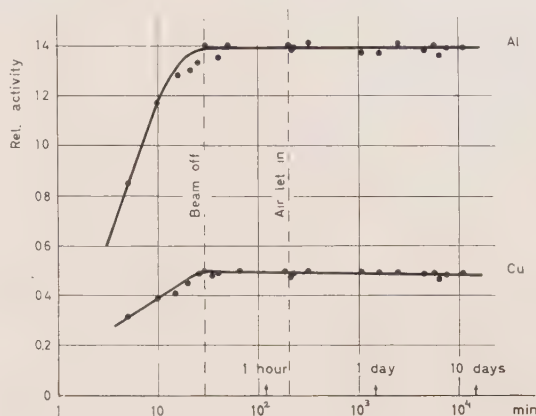


Fig. 14. Time stability of Kr targets.

lected in Al is absolutely constant during the time of experiment, but in Cu there is a slight decrease. Other soft materials such as Pb and Tl have shown the same property as Cu, while a graphite target, which is fairly porous, did not change its activity at

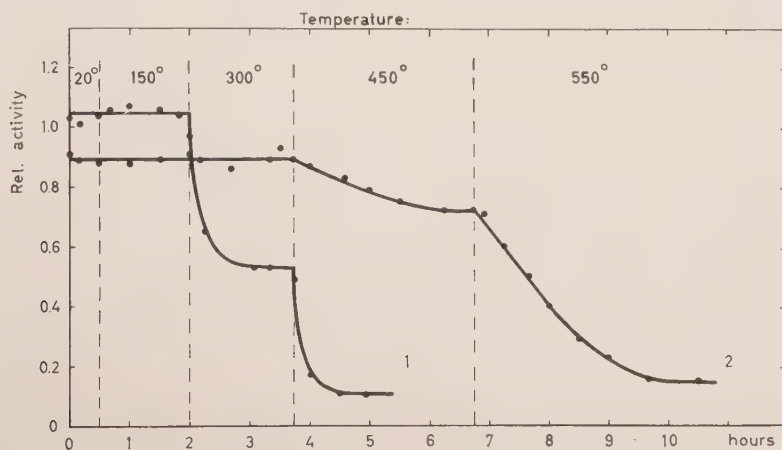


Fig. 15. Temperature stability of Kr collected in Ag. Foil 2 was annealed.

tion chamber of the separator. The activity was compared with a Ta target containing Kr, which was aged for several months. Since we expect an eventual change of collected gas to be large during the first hours and then decrease, we may regard this Ta target as practically constant for an ad-

all. Experiments with Xe gave the same result, and it is our impression from measurements with a high voltage machine that Ne and Ar stored in Mo and Ta backings are also very permanent. Experiments with Ne in Cu, on the other hand, have shown a very fast decrease, so that all Ne atoms have left

within a few days. It seems to be the formation of porous oxide on the surface, which causes the decrease of collected gas in Cu, Pb and Tl, rather than a diffusion. If diffusion at room temperature were of the order measured here, there should be an appreciable decrease of Kr at a slightly increased temperature, which is not the case (see below).

Fig. 15 illustrates the effect of a temperature increase on an Ag foil containing Kr. Studies have been made both for a normal rolled Ag foil (1) and for a foil which had been heated in a vacuum to a high temperature just before use (foil 2). The difference at room temperature, about 10 per cent, is reproducible and already mentioned in § 3.3. From fig. 15 it is seen that at 300°C there is a fast diffusion in foil 1, but no detectable diffusion in foil 2. At about 450°C diffusion starts also in this material, but the decrease is only about 15 per cent. Heating to 550°C means a strong outgassing, so that after a while 15 per cent remains in foil 2. The difference in temperature stability between the two qualities may be caused by the stronger annealing process in the rolled material, which results in a higher diffusion. On the other hand, it is surprising that the saturation value at room temperature is higher in rolled material.

Cu gave about the same characteristic curve, but the decrease was not so fast, and there was no difference whether we had vacuum heated the Cu foils or not. At 550°C about 60 per cent of the gas remained in the foil. In materials such as Mo and Ta only a slight decrease of Kr took place below 600°C. In Ni and Pt nearly 90 per cent remained at 600°C. No difference was found between rolled material of Ni, Mo and Ta, and material heated in vacuum, but this indicates only that heating does not change the crystal structure as fast as in Ag.

Noble gas targets on many backing materials are thus very stable, regarding both time and temperature. Normal use in high voltage machines is therefore not expected to destroy gas targets prepared on suitable backings.

8. Sputtering Experiments

8.1. MEASURING METHOD

Very soon it became clear that the saturation process depends not only on diffusion of gas atoms

but also on the sputtering rate of the backing material. Therefore, we started an investigation of sputtering phenomena, bombarding with noble gas ions many of the backing materials for which saturation values were already known.

The sputtering ratio S is given as the number of atoms of the backing material thrown out for every incoming ion. The target foil must be weighed before and after bombardment. This was done by means of a micro-balance. The current I (in μA) was measured in a Faraday cage as is indicated in

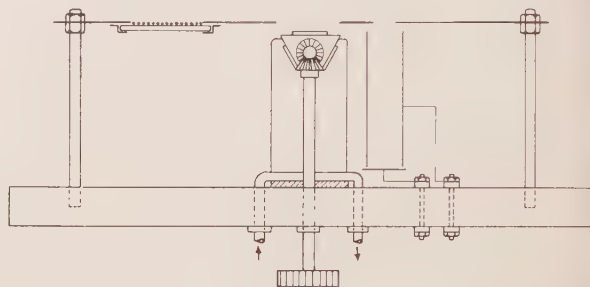


Fig. 16. Target arrangement for sputtering measurements.

fig. 16. Three targets mounted on a turnable triangular body could be bombarded successively. This arrangement simplified our measurements a great deal. Since saturation values are very small for noble gases, a few $\mu\text{g}/\text{cm}^2$, the weight of collected atoms is negligible compared with Δm μg , the weight of the material sputtered off. Therefore we get the sputtering ratio as

$$S = 26.6 \frac{\Delta m}{M_2 I t} \text{ atoms/ion} \quad (5)$$

where M_2 = mass number of investigated material;
 t = time of bombardment in hours.

We have seen above that materials of different pre-treatment can store slightly different amounts at saturation, and it is expected that sputtering also can be different for the same reason. Fig. 17 shows an investigation of the change of S , when more and more material is sputtered off, using Kr ions of 45 keV energy and 15 $\mu\text{A}/\text{cm}^2$. Two different curves 1 and 2 are given for two commercial qualities of Ag. The upper curve, 2, approximately coincides with the curve which we get for Ag heated in vacuum before use. From fig. 17 we also see that S may be

by as much as 25 per cent higher if $2000 \mu\text{g}/\text{cm}^2$ is sputtered off from a new foil instead of $200 \mu\text{g}/\text{cm}^2$. Above $1500\text{--}2000 \mu\text{g}/\text{cm}^2$ S is approximately independent of the sputtered amount. If comparable results are wanted we must either use new foils and sputter off the same amount in each experiment, or use foils from which a large amount of material has been removed. When an Ag target is used many times for gas ion bombardment this starting up period is thus avoided. In sputtering experiments described in the following, the quality 1 of Ag is used.

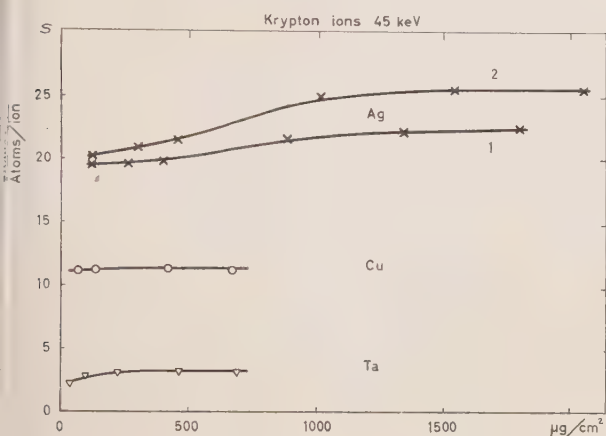


Fig. 17. Change of sputtering ratio with the amount of removed material. Still higher sputtering ratios have been measured for another quality of Ag: 30–32 atoms/ion.

In the case of Cu and Ta we see that at least $200 \mu\text{g}/\text{cm}^2$ must be sputtered away to get a sputtering ratio independent of the removed quantity. However, the increase of S is not as serious here as it is for Ag. The change of the sputtering ratios with quantity has not been investigated systematically for other materials, but there is evidence that variations of a few per cent are common.

The reason for the increase can be that an oxide layer on the surface protects the underlaying material. When this layer is removed, sputtering increases. Another reason is the increase in surface area caused by particularly intensive removal of material at crystal boundaries. That sputtering is higher at the boundaries can be observed in a microscope after bombardment. At the same time the angle of beam incidence is also changed to a mean value different from a perpendicular incidence,

when the surface is eroded, and we shall see later that sputtering increases, when the angle of incidence is increased.

In all experiments described here we sputtered off more than $200 \mu\text{g}/\text{cm}^2$. The area of the beam was about 1 cm^2 , and current density about $100 \mu\text{A}/\text{cm}^2$. The pressure in the target chamber was $(0.8\text{--}1.5) \times 10^{-5} \text{ mm Hg}$. The accuracy of the micro-balance was better than $\pm 5 \mu\text{g}$ in a careful weighing. The error of the current measurement was less than ± 5 per cent. Therefore, sputtering ratios may be determined with an accuracy of about 7 per cent, when the same material is used. As stated above different qualities of a material may have slightly different sputtering ratios. For this reason many results given here must be regarded as mean values and variations of 7 to 15 per cent may occur.

8.2. DIFFERENT TARGET MATERIALS

In fig. 18 sputtering ratios for Kr ions of 45 keV on some 30 materials are shown in a diagram similar to fig. 6. We find that S varies within the same periods of the periodic table as those pointed out in the discussion of fig. 6. A comparison between these two diagrams shows that a low sputtering ratio always means a high saturation value and vice versa. The only exception is Sb where the sputtering ratio is higher than expected from the measured

TABLE 5
Sputtering ratios for Ne, Ar, Kr and Xe ions of 45 keV on different materials.

Target material	Sputtering ratios atoms/ion				
	Ion:	Ne	Ar	Kr	Xe
V		0.3	1.0	1.7	1.9
Fe		1.3	2.3	4.0	4.9
Ni		1.4	3.5	5.6	7.6
Cu		3.2	6.8	11.8	19.0
Mo		0.6	1.5	2.7	3.8
Pd		2.5	5.3	10.5	14.4
Ag		4.5	10.8	23.5	36.2
Sn		1.8	4.3	8.5	11.8
Ta		0.7	1.6	3.1	4.0
W		1.0	2.3	4.7	6.4
Pt		1.9	5.3	11.3	16.0
Au		3.6	10.2	24.5	39.0
Pb		3.6	10.5	24.0	44.5

saturation. However, the Sb used in the measurements had a very coarse structure, and it is likely that this is the reason why the observed irregularity occurs.

Sputtering ratios for Xe ions are somewhat higher than those for Kr and the variations with

piece of good quality graphite. Vaporized carbon foils must have somewhat lower sputtering, since the saturation value is much higher in such foils than in block material. Our measurements on Cr and Rh electrolyzed on Cu and Ag, respectively, gave sputtering ratios, which were lower than those

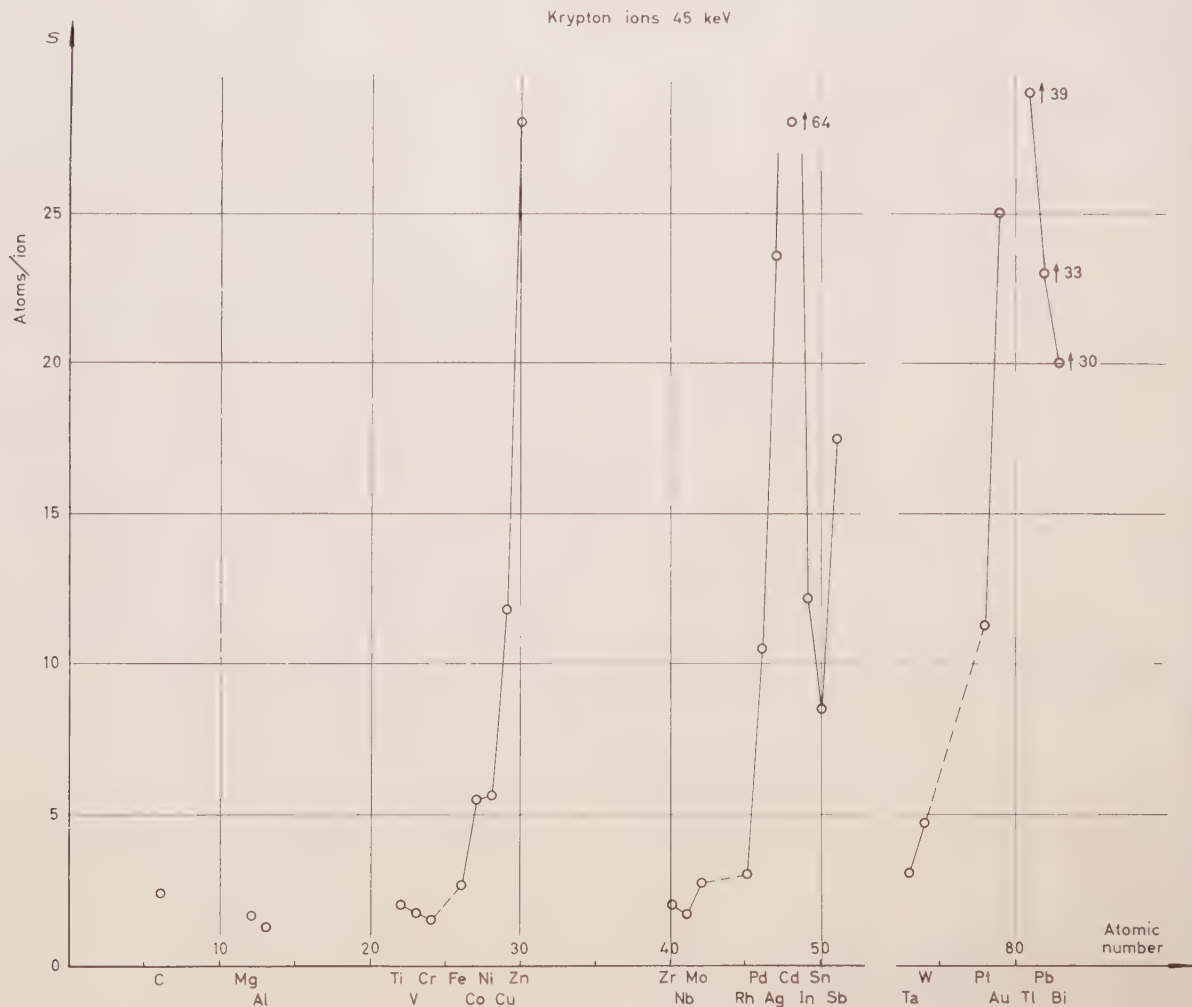


Fig. 18. Sputtering ratios for 45 keV Kr ions bombarding different target materials.

target material are similar. Therefore, only Kr sputtering is represented in fig. 18. (Not shown in this figure is the sputtering of Hf, 2.5 atoms/ion.) Instead, sputtering ratios are given in table 5 for 13 different materials bombarded with Ne, Ar, Kr and Xe ions of 45 keV.

As target materials in most cases we used commercially available foils as already mentioned in § 4. The C sputtering in fig. 18 was measured on a

expected for a crystal structure corresponding to ordinary rolled material. This is concluded from a comparison of the groups Cr, Mo, W and Co, Rh with groups such as Ni, Pd, Pt and Cu, Ag, Au which show a regular systematics.

All sputtering targets were handled carefully and cleaned with ether before first use. On the other hand no attempt was made to smoothe the surface, because even the smoothest surface is fairly rough,

when the penetration depth of the impinging particles is taken as a scale. The target was water-cooled and temperatures very seldom exceeded 100°C. When materials with low melting points, e.g. Sn and In, were used, the beam had to be swept over the target in order to decrease the current density. Otherwise the foil was burned through.

3. SPUTTERING RATIOS VERSUS ION ENERGY

Figs. 19a and 19b show the sputtering ratio as a function of ion energy for noble gas ions Ne, Ar, Kr and Xe, bombarding on Cu and Ag. Sputtering for

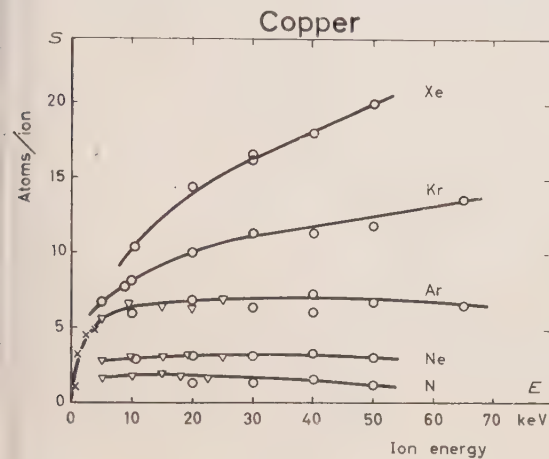


Fig. 19a.

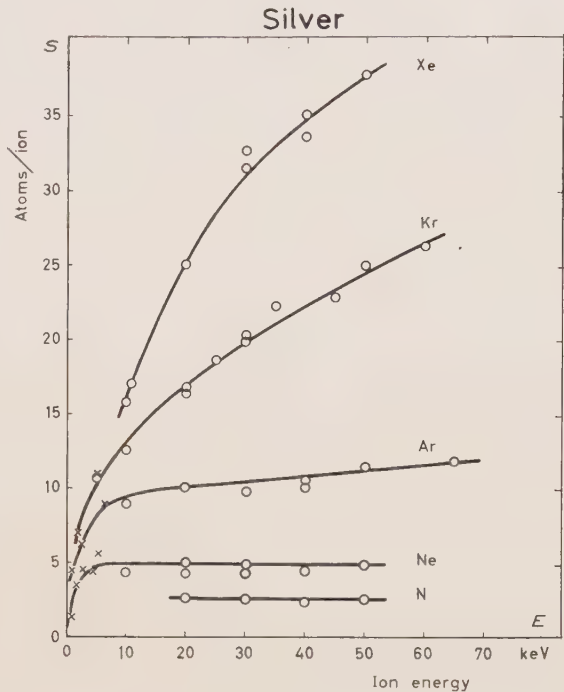


Fig. 19b.

Fig. 19. Sputtering ratios versus ion energy.

N ions is also indicated in the diagrams. We note that for N, Ne and Ar ions, S is fairly constant, when the ion energy is higher than 5 keV, corresponding to a penetration depth of about $10 \mu\text{g}/\text{cm}^2$. S even decreases slightly for Cu. For heavier ions sputtering increases with energy within the whole energy range. However, a maximum may be expected for a sufficiently high ion energy. In the sputtering diagrams sputtering ratios determined by Rol¹⁰) (∇) and by Keywell⁸) ($\times$) are indicated and the agreement with our measurements is very good.

In figs. 20a, 20b and 21 sputtering ratios for some other materials are given. Here we observe that S becomes constant above ion energies varying from 10 to 30 keV. On Sn we got a considerably decreasing sputtering ratio with increasing ion energy until we detected that in this case S also depends on

the power dissipated in the Sn foil. When we used the same power throughout the experiment we measured the sputtering given in fig. 20b.

It is interesting to compare fig. 19b with figs. 20a, 20b and also with fig. 21. All the target materials

have practically the same mass number M_2 , about 100. Not only does the sputtering ratio vary considerably for different elements, but the shapes of the curves also are quite different. This indicates a complicated influence of the properties of individual elements on sputtering. A complete theory of sputtering must be able to explain all features indicated here. However, as to be shown in § 9 it is possible to calculate sputtering ratios approximately for noble gas ions bombarding on solids.

8.4. CURRENT DENSITY

For Cu and Ag we have measured the sputtering ratio at different current densities of 45 keV Kr ions. The metal foils used were first bombarded at high current densities to remove the surface layer. After that, three targets were bombarded using the mass numbers 82, 84 and 86, respectively. If we

assume the focusing of the ion beam to be approximately the same for all mass lines, the current densities are proportional to the isotopic abundances. Approximately equal amounts were sputtered off from all targets (cf §8.1). The result was

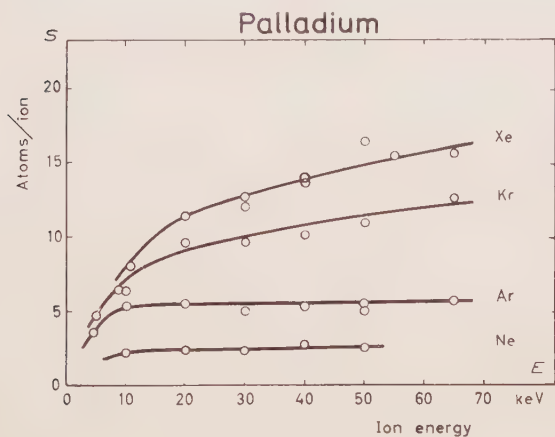


Fig. 20a.

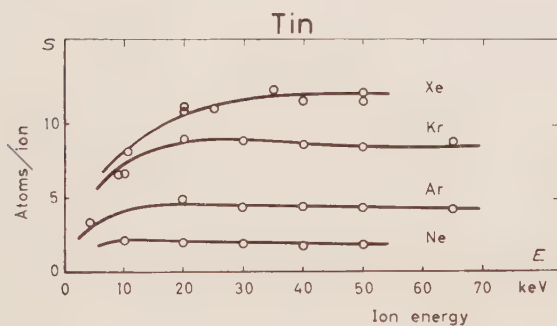


Fig. 20b.

Fig. 20. Sputtering ratios versus ion energy.

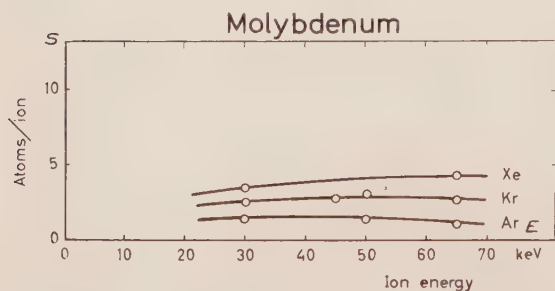


Fig. 21. Sputtering ratios versus ion energy.

that the sputtering ratio is independent of current density between 10–100 $\mu\text{A}/\text{cm}^2$. This might not hold for much higher current densities due to the high power dissipation in a thin layer, and the accompanying changes of crystal structure (§ 8.7).

8.5. VACUUM IN THE TARGET CHAMBER

Fig. 22 shows the variation of the sputtering ratio with pressure and composition of the rest gas in the target chamber. Kr ions of 45 keV were used. A liquid air trap near the target was used to minimize the effect of gaseous products from the vacuum oil. As can be seen the Ar gas has practically no influence at pressures below 10^{-4} mm Hg. Oxygen, on the other hand, decreases the sputtering ratio of Cu and Ta considerably, (cf ref.¹³), while the sputtering of Ag is not influenced.

Since the ion deflection chamber of the separator is quite narrow, the pressure in the target chamber

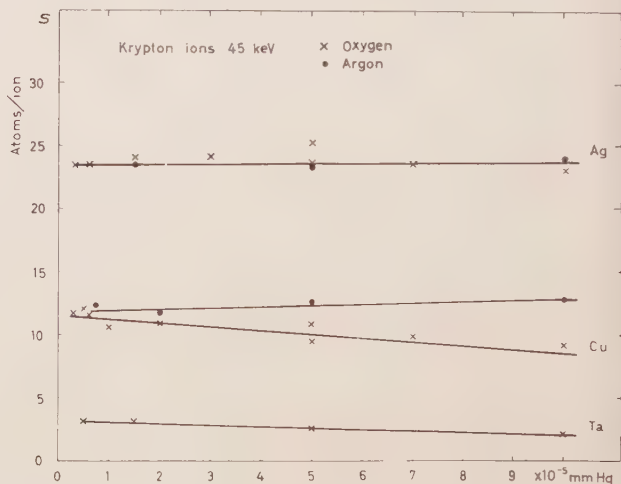


Fig. 22.

Change of sputtering ratio with pressure in the target chamber.

is hardly dependent on pressure fluctuations in the ion source compartment. A large diffusion pump at the target end of the machine produces a constant vacuum well within the limits $0.8\text{--}1.5 \times 10^{-5}$ mm Hg. The variations due to pressure changes are thus small in our sputtering measurements.

8.6. ANGLE OF ION BEAM INCIDENCE

In some cases we have measured the sputtering ratio at different angles between the ion beam and the target plate. The result is given in figs. 23a and b for 45 keV Kr ions and 45 keV Ne ions, respectively. We see that the increase of the sputtering ratio is considerable so that at 60° S is greater by a factor of two than at 0° . No simple equation can

¹³ O. C. Yonts, J. Appl. Phys. 31 (1960) 1583.

describe the variation of S with the angle of incidence, although it is possible to fit expressions to describe the individual curves. E.g.

$$S_{\varphi} \approx S_0 \frac{1}{\cos^{\frac{1}{2}} \varphi} \quad (6)$$

where

S_{φ} = sputtering ratio at φ° angle of incidence;

S_0 = sputtering ratio at perpendicular incidence,

fits the curves for Cu, Ni and Ta fairly well between $0-60^\circ$, but cannot explain that for Ag.

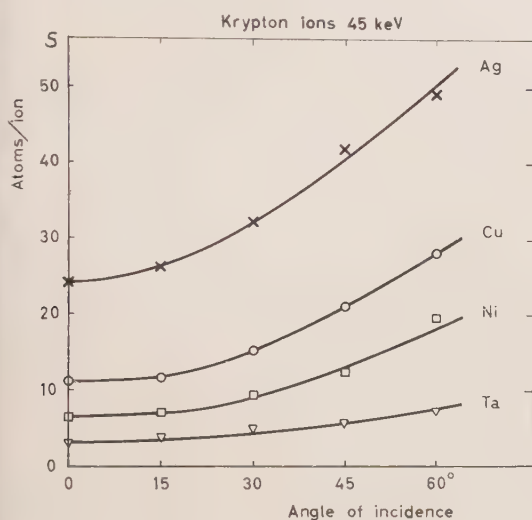


Fig. 23a.

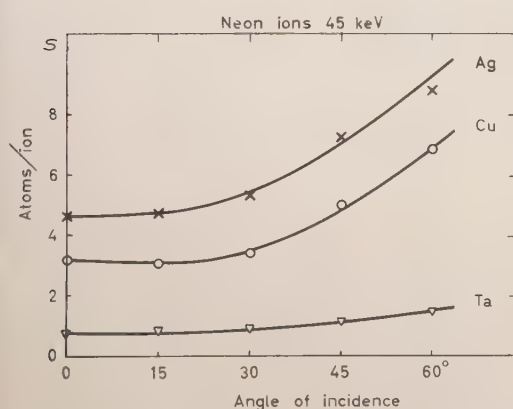


Fig. 23b.

Fig. 23. Sputtering ratio versus angle of ion beam incidence.

8.7. TARGET TEMPERATURE

The sputtering experiments described so far were performed at a low temperature of the bombarded material, less than 100°C . In the experiments on

Sn we found an influence of the power dissipated on the foil as mentioned above and this indicated that an increased temperature may decrease the sputtering ratio.

Fig. 24 reproduces measurements on three backing materials. Kr ions of 45 keV were used. S is constant for Ag up to 600°C , but increases at higher temperatures due to vaporization losses. For Ni and Pt, on the other hand, S decreases slowly when the temperature is raised. It is to be

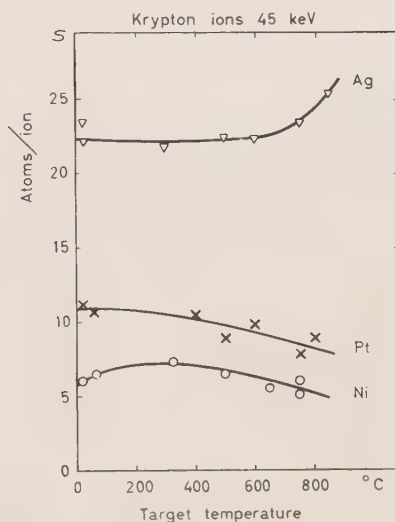


Fig. 24. Sputtering ratio versus target temperature.

expected that S will start to increase at some still higher temperature, but our experimental arrangement did not permit higher temperatures than 800°C . The decrease of S must be due to annealing of the metal at high temperatures, so that the damage caused by one entering ion is more or less eliminated before a new ion hits close to the spot of incidence of the first one. In most materials the influence of temperature on the sputtering ratio is negligible below $200-300^\circ\text{C}$.

9. Discussion of Sputtering Phenomena

Several authors have tried to calculate sputtering ratios from ion energy and mass numbers of ion and target atoms. Most of the work is limited to the sputtering of very light ions, ($M_1 = 1-4$), but there are attempts to estimate the ratio also for heavy ions.

Keywell⁸⁾ has thus used a modified neutron

cooling theory, to calculate how the impinging particles give off their energy in successive collisions with the target atoms. The chance for displaced atoms to leave the material is also taken into account. This theory may fairly well explain the measured sputtering ratios at low ion energies, below 5 keV. For higher energies the theory fails in most cases. Especially, it cannot explain the decrease of sputtering which is observed for light ions, e.g. Ne ions, at high energies.

E = energy of impinging particle;
 M_1, M_2 = mass numbers of ion and target atoms;
 K = a constant.

The mean free path is given by the collision diameter R and the number of atoms per unit volume of the target material n_0

$$\lambda = \frac{1}{\pi R^2 n_0} \quad (8)$$

The collision diameter is calculated by assuming

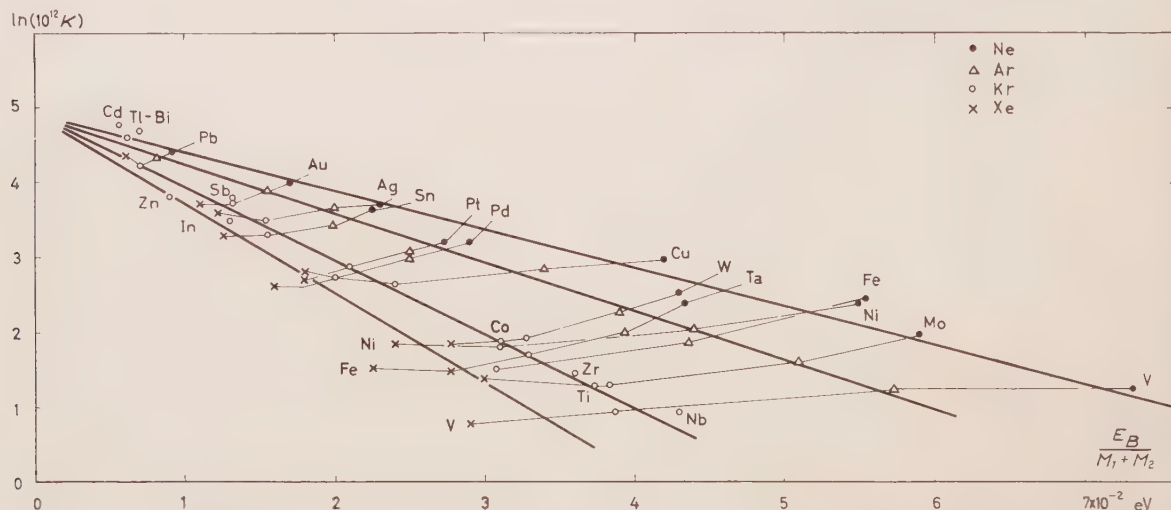


Fig. 25. $\ln K$ versus binding energy of target atoms divided by the sum of ion and target mass numbers. For Pd better agreement with the straight lines is obtained, if $E_B = 4.04$ eV (Brewer) is used instead of 3.64 eV.

Another theory, given by v. Hippel¹⁴) explains qualitatively the sputtering as vaporization from heated spikes developed around the spots, where individual ions enter the material. However, it is difficult to use this theory for direct calculations, although some properties of the sputtering process might probably be explained by the theory.

Rol¹⁰) has recently calculated sputtering ratios using fundamental equations given by Bohr¹⁵). Rol assumes that the sputtering ratio is proportional to the energy dissipated by the impinging particle in the first atomic layers. The chance that a collision takes place near the surface is inversely proportional to the mean free path λ , and the sputtering can be written

$$S = K \frac{E}{\lambda} \frac{M_1 M_2}{(M_1 + M_2)^2} \quad \text{atoms/ion} \quad (7)$$

that the slowing down of an ion takes place in a screened coulomb field of a type proposed by Bohr.

$$R = \frac{a_0}{(Z_1^{\frac{2}{3}} + Z_2^{\frac{2}{3}})^{\frac{1}{2}}} \ln \frac{Z_1 Z_2 e^2 (M_1 + M_2)}{4.7 \epsilon_0 R E M_2} \quad (9)$$

where

Z_1, Z_2 = atomic numbers of ion and target atom;
 e = charge of an electron;
 ϵ_0 = dielectric constant for vacuum;
 a_0 = radius of the first electron orbit in a hydrogen atom. MKS units are used.

We have investigated the energy dependence of sputtering for more than 40 combinations of ion and target material. Some of the results are shown in figs. 19–21. By properly adjusting the K -values, nearly all curves are found to fit eq. (7) for the whole investigated energy range (10–65 keV). Thus all our measured sputtering ratios (as function of

¹⁴) A. v. Hippel, Ann. der Physik **80** (1926) 672.

¹⁵) N. Bohr, Mat. Fys. Medd. Dan. Vid. Selsk **18**, no. 8 (1948).

energy as well as at 45 keV) could be used to calculate K -values. These values were plotted against different parameters (mainly physical properties of the material), and we found that $\ln K$ as a function of $E_B/(M_1 + M_2)$ gives straight lines in a diagram (fig. 25), one for each noble gas. E_B is the binding energy of the target atoms. The slopes of these lines are proportional to $-\sqrt{M_1}$ and therefore we can write

$$K = a \cdot \exp \left(\frac{-b \sqrt{M_1}}{M_1 + M_2} E_B \right) \quad (10)$$

a and b are constants.

In fig. 25 we see that there are deviations from the proposed straight lines greater than might be explained by our experimental error and by the uncertainty of the binding energies.

It is reasonable to assume that the number of target atoms displaced at the first collision of an entering ion is proportional to $KEM_1M_2/(M_1 + M_2)^2$. The probability for the collision to take place in the surface layer is proportional to $1/\lambda$. However, it is not sure that the sputtered fraction of these displaced atoms becomes equal for all different materials. Instead it must be assumed that this fraction depends on the mean free path of the displaced particles and on the properties of the metal lattice. The mean free path could be calculated from eq. (8), if the energy of the displaced atoms were known. Of course there is a distribution with widely spread energies, but probably the mean energy could be used for calculations. In this case more experimental data are needed before the sputtering process can be completely explained.

A few preliminary measurements have been made to determine the mean energy of the sputtered particles. A thin copper plate was suspended from two thin wires, one of copper and one of constantan, thus forming a thermocouple. With this arrangement placed in front of the sputtering target, we could measure the energy of the atoms which were sputtered out perpendicularly from the target. The incidence angle of the ion beam was 45° . We got the mean energies given in table 6, where Kr ions of 45 keV were used. Variations with ion energy were very small between 25–50 keV. The given values are somewhat too high, since all impinging atoms do not stick on the collector, the self-sputtering ratio at these energies not being negligible.

In a similar way we have also measured the momentum transferred by sputtered Ag atoms. From this we have calculated the mean velocities 7.1, 6.3 and 4.1 10^3 m/s, when Ar, Kr and Xe ions of 45 keV were used.

TABLE 6

Target material	Energy of sp. atoms (eV)
Cu	45
Ag	36
Ta	250
Au	60

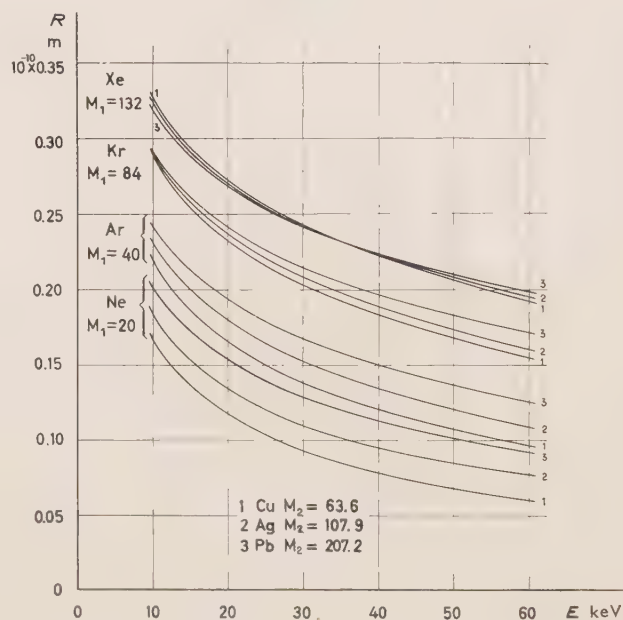


Fig. 26. Collision diameters calculated[†] for noble gas ions bombarding Cu, Ag and Pb.

If we use eqs. (7), (8) and (10) with numerical values for the constants a and b , we get a final expression for sputtering with noble gas ions.

$$S \approx 4.24 \times 10^{-10} n_0 R^2 E \frac{M_1 M_2}{(M_1 + M_2)^2} \exp \left(-10.4 \frac{\sqrt{M_1}}{M_1 + M_2} E_B \right) \text{ atoms/ion} \quad (11)$$

(E_B and E in eV, M_2 mass number, R in m and n_0 in m^{-3} .)

For E_B we have used data, compiled in Thermodynamic Properties of Metals and Alloys¹⁶⁾ or, where missing in these tables, the data given by Brewer¹⁷⁾.

[†] R is calculated by means of Wegematic 1000 at the ADB Institute, Chalmers University of Technology.

¹⁶⁾ Thermodynamic Properties of Metals and Alloys, Minerals Research Laboratory, Univ. of Cal. Berkeley.

¹⁷⁾ L. Brewer, Univ. of California Radiation Laboratory, Berkeley UCRL 2854 (rev. 1955).

To simplify the use of eq. (11) R has been computed for a number of target materials bombarded by noble gas ions of different energy. Some results are given in fig. 26. Values between two chosen mass numbers can easily be interpolated. The probable deviation between eq. (11) and all

orientation of the crystal. Therefore it might be expected that the saturation value also should vary with the orientation.

In fig. 27 the saturation value is represented as a function of the angle between the normal to the surface of a Cu single crystal and the direction of

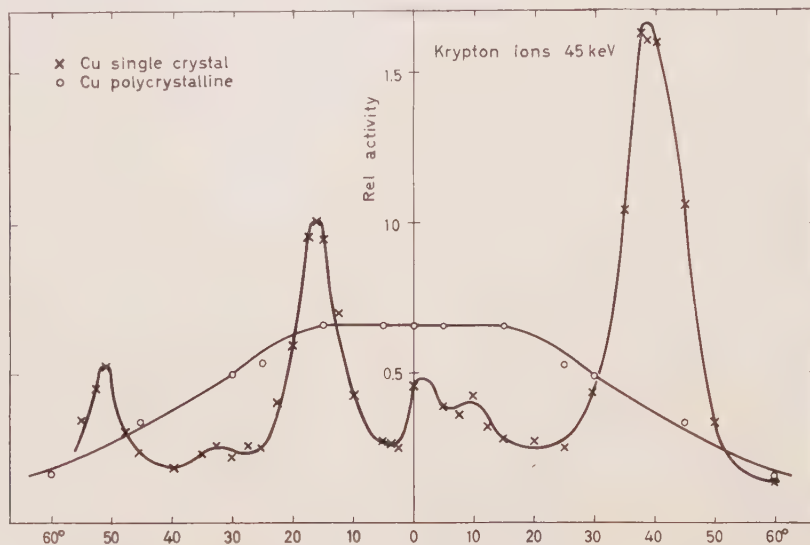


Fig. 27. Saturation value in a Cu single crystal turned around an $[110]$ axis in the (111) plane. The $[111]$ direction is 3° from zero.

our experimental results is about 20 per cent. The greatest deviations are found for Mg and Al not included in fig. 25, but for these metals a thick oxide layer decreases the sputtering ratio.

Sputtering ratios for most other ions cannot be calculated by means of this simple formula for noble gas ions, because saturation values are in many cases no longer negligible. Instead, the collected atoms change the properties of the undisturbed material considerably. The measured sputtering is thus no longer representative for the target material alone, and it also depends on operating conditions, such as time of bombardment, current density and target temperature. These phenomena will be discussed in a following paper.

10. Collection in and Sputtering of Single Crystals

According to Rol *et al.*¹⁸⁾ the sputtering ratio for ions bombarding a single crystal depends on the

the ion beam. The orientation in the surface was determined from a Laue diffraction pattern to be (111) with an error of $\pm 2^\circ$. The crystal was mounted in an arrangement similar to that in fig. 1, but constructed so that the crystal could be rotated round an $[110]$ axis in this (111) plane. We collected the same mixture of Kr^{84} and Kr^{85} as used previously (§ 3) until the crystal was saturated. The ion energy was 45 keV and current density $10 \mu\text{A}/\text{cm}^2$. After saturation the activity was compared with the activity of a reference. The ratio between these two is reproduced in fig. 27. The Kr already collected was sputtered off by means of inactive Kr ions before investigating the next setting of the entrance angle. It was observed that the collected activity could not be completely removed by sputtering, but we tried to sputter off to approximately the same rest activity. This was taken as background, and subtracted from each measured value. As a comparison fig. 27 also shows the collection curve for polycrystalline material at different angles of incidence.

¹⁸⁾ P. K. Rol, J. M. Fluit, F. P. Viehböck and M. de Jong, Proc. Fourth Conf. on Ionisation Phenomena in Gases, Uppsala 1959 (North-Holland Publ. Co., Amsterdam, 1959) 257.

Since saturation and sputtering are related, we have also measured the sputtering ratio for Kr ions of 45 keV bombarding a Cu single crystal. The result is shown in fig. 28, where sputtering of polycrystalline Cu also is given for different angles of incidence. The single crystal retained its appearance throughout the series of experiments, although

made to find out definitely the relation between the three parameters discussed here.

In the diagrams transparent directions turn up, e.g. from the right $[110]$ $35^\circ .3$, $[233]$ $10^\circ .0$, $[111]$ 0° , $[112]$ $19^\circ .5$, $[114]$ $35^\circ .3$ and $[100]$ $54^\circ .7$. Still higher indices cannot be detected in our investigation, since the intervals between the measuring

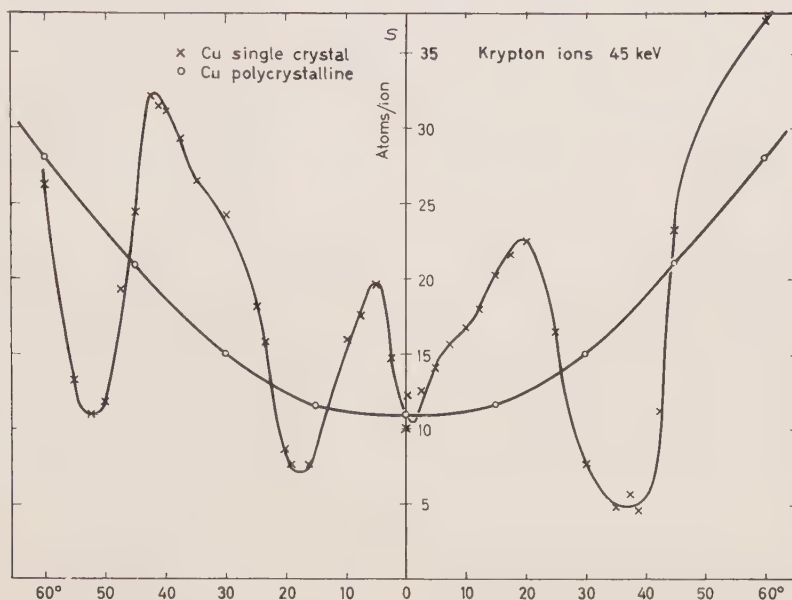


Fig. 28. Sputtering ratio for a Cu single crystal turned around an $[110]$ axis in the (111) plane. The $[111]$ direction is 3° from zero.

the crystal was heavily bombarded by energetic ions. The reason is that the displaced atoms after slowing down can be fixed again to vacant lattice sites, and also that the damaged layer is continuously sputtered off.

From a comparison of figs. 27 and 28 we see that a high saturation value is combined with a low sputtering ratio and vice versa. At angles of incidence with high saturation the crystal is more transparent than at other angles, which indicates that the penetration depth must vary in the same way as the saturation value. Since diffusion also varies with the transparency of the crystal, the penetration depth cannot be proportional to the saturation value. However, the sputtering ratio is fairly high and influences the saturation more than diffusion does. This means that the penetration depth should be approximately proportional to the saturation value. Further experiments must be

points are too large. It is also to be noticed that the ratio between the maxima of single crystal sputtering and the corresponding sputtering of polycrystalline material is nearly constant. In the same way there is a constant ratio between the minima of saturation in the single crystal and of the saturation in polycrystalline material.

Some preliminary saturation measurements have also been made for Al single crystals. These crystals were not of the same high quality as the Cu crystal. However, the measurements indicate that also in a low sputtering metal, the saturation value depends on the orientation of the crystal. In fig. 11b the saturation value versus ion energy is about 20 per cent higher for a (110) plane in the surface than for polycrystalline material. From the measurements on Cu we would expect a greater difference. Probably either the oxide layer on Al, or the higher Kr concentration in Al compared with

that in Cu, suppresses the amplitude of the curve in fig. 27.

11. Discussion of the Collection Process

We can now use eqs. (1) and (2) to discuss the collection process. The distribution of trapped particles within the target material at saturation is hitherto known only in a few cases. The (p, γ) resonance curve in fig. 3 represents approximately the distribution of Ar atoms in Ta and Au. Davies *et al.*¹⁹⁾ have measured distributions of Cs and Na ions in Al. The concentration of trapped particles was here far below saturation. From these experiments it seems obvious that the distribution curve has a different shape in different materials. In a material where diffusion is high, we also expect a low concentration near the surface. This means that no simple relation between f_s and $\bar{f}$ can suit all cases of collection.

For an approximate calculation we assume,

1. the mean concentration of trapped atoms near the surface, f_s , is about half the mean concentration within the penetration depth $\bar{f}$,

2. the penetration depth is twice that calculated from Nielsen¹²⁾ (see refs.¹⁹⁾ and ²⁰⁾).

With these assumptions we obtain a diffusion ratio at saturation, which is near 1 for low sputtering materials Al, Ti, Zr, Nb etc, and near zero for high sputtering materials such as Ag, Cd, Au and Pb. This result is reasonable and shows that

our assumptions may be approximately right. Since the uncertainty of the calculation is too large, because of unknown parameters, we will not present more detailed results here. Also, it has not been possible to explain the observed diffusion by means of known properties of the target material.

Our conclusion is, however, that the saturation process depends on two main parameters (i) sputtering, which is determined by the binding energy of the target atoms and the mean free path of displaced target atoms, (ii) diffusion of gas atoms. The latter requires more experimental work for a quantitative explanation.

Acknowledgements

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The Cu single crystal belongs to Philips Research Laboratories, Holland, and was placed to our disposal by Dr. P. K. Rol.

¹⁹⁾ J. A. Davies, J. D. McIntyre, R. L. Cushing and M. Lounsbury, Can. J. Chem. **38** (1960) 1535.

²⁰⁾ K. O. Nielsen, Thesis (Copenhagen, 1958).

SPUTTERING EXPERIMENTS IN THE HIGH ENERGY REGION

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Sputtering ratios for three target metals, Cu, Ag and Ta, have been determined for 45 keV ions of some 70 elements. A formula for estimation of the collected amounts of ions in a target material is given. From this the amounts collected in Ta are

calculated and compared with experimental determinations. In addition selfsputtering ratios have been determined for about 25 elements at 45 keV ion energy, and for some of them also at lower energies.

1. Introduction

In a preceding paper¹⁾ we have studied saturation and sputtering phenomena for noble gas ions bombarding on different materials. This work was performed in order to produce high quality noble gas targets for (p, γ) resonance studies in high voltage machines.

Of course it is also of great interest to study the collection and sputtering of other ions than of noble gases. The tracer technique is not so easily adapted to ions of solid substances as it is for gases. Therefore we have measured sputtering ratios and from the results estimated the collected quantities.

The electromagnetic isotope separator²⁾ at our laboratory has been used for all our experiments. This means that we have used an analyzed ion beam for measuring sputtering ratios.

2. Experimental Determination of Sputtering Ratios

The same target arrangement was used here as in the previous investigation¹⁾. On a revolvable triangular body three targets can be mounted and bombarded successively. The copper holder can be cooled by water in order to prevent the target plate to be heated too much, when high current densities are used. The ion current I was measured with a Faraday cage placed beside the sputtering target. The target plate was weighed on a micro-balance before and after bombardment.

The measured decrease in weight of the target plate is called Δm . This is the difference between sputtered quantity of target material, Δm_2 , and increase in collected amount of ions, Δm_1 . The sputtering ratio can therefore be written

$$S = 26.6 \frac{\Delta m + \Delta m_1}{M_2 \cdot I \cdot t} \text{ atoms/ion} \quad (1)$$

(Δm and Δm_1 in μg , I in μA , M_2 mass number of target atoms and t time in hours).

If we bombard until saturation of bombarding particles is reached, which means that $m_1 = \text{constant}$, and afterwards measure the sputtering ratio, Δm_1 is equal to zero and

$$S = 26.6 \frac{\Delta m}{M_2 \cdot I \cdot t} \text{ atoms/ion} \quad (2)$$

This equation is also approximately valid when $\Delta m_1 \ll \Delta m$.

We can also bombard the target material M_2 with ions of the same element. We assume that a fraction η_i of the incoming ions is trapped in the target material. If the loss of weight is Δm , the selfsputtering ratio is

$$S_e = 26.6 \frac{\Delta m}{M_2 \cdot I \cdot t} + \eta_i \text{ atoms/ion} \quad (3)$$

In most cases η_i is 1. This is valid at least for all low sputtering metals. For high sputtering metals η_i gives only a small contribution to S_e , and therefore we can assume η_i always to be 1. We observe that Δm can be positive or negative, which means a sputtering ratio above or below 1.

¹⁾ O. Almén and G. Bruce, Nucl. Instr. and Meth. 11 (1961) 257.

²⁾ O. Almén, G. Bruce and A. Lundén, Nucl. Instr. 2 (1958)

For some investigations it is necessary to have the same current density over the whole target. In such cases we modulate the ion beam by means of a saw-tooth voltage on two deflection plates, placed at the entrance side of the separator magnet (see ref. 1)).

high quality insulators, carefully protected from the plasma. Where suitable chemical compounds do not exist, and the elements themselves have a low vapour pressure, the elements have been handled in a sputtering ion source. Concerning ion source problems see ref. 3, 4).

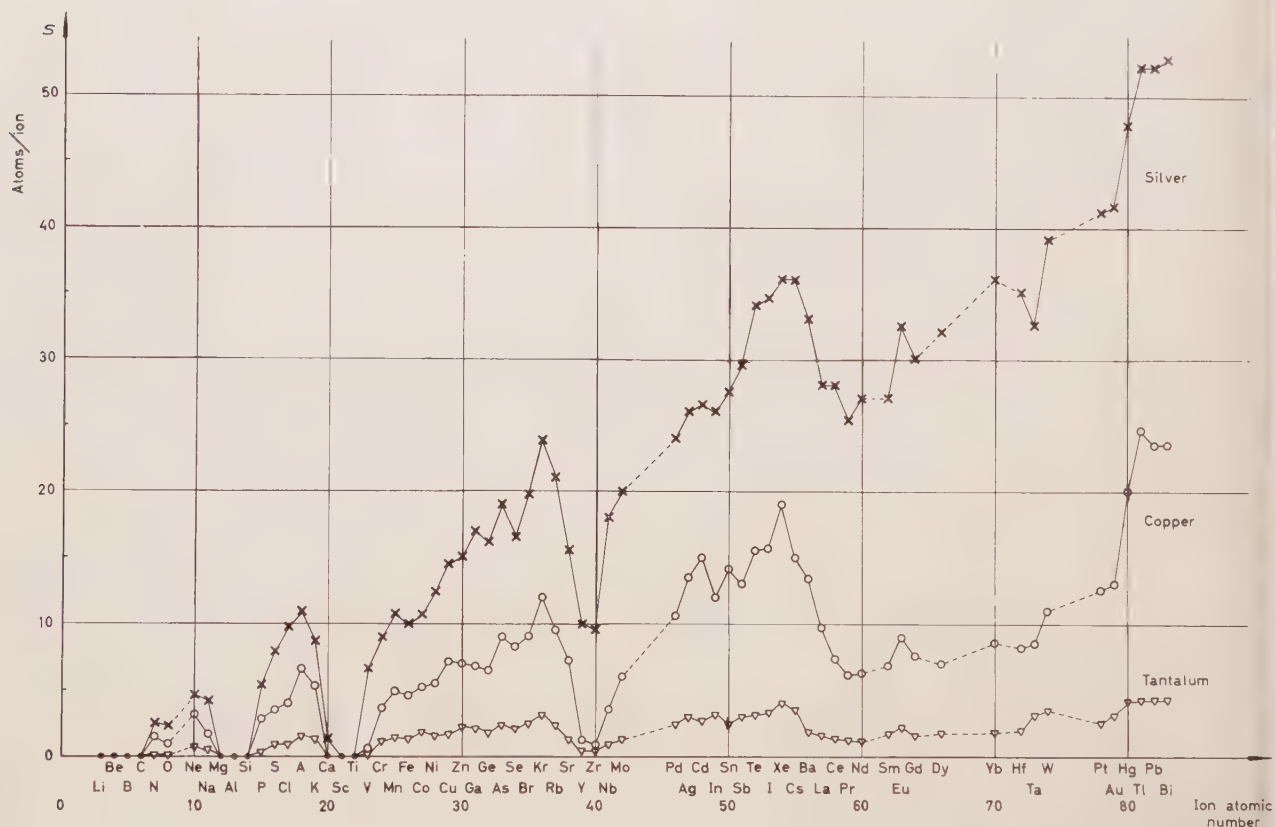


Fig. 1. Sputtering ratios for different ions of 45 keV. (In order to get comparable values, S_e-1 is plotted instead of the three selfsputtering ratios.)

The handling of many different elements for the production of ion beams requires the use of different types of ion sources. All gaseous elements, and pure elements with a vapour pressure of more than 10^{-2} mm Hg at a temperature lower than about 500°C can normally be handled in a simple low temperature ion source. In many cases chemical compounds must be used. Chlorides are often the most suitable charge materials. The higher the required temperature of operation, the more intense is the chemical attack on ceramic materials used as insulators in the ion source. Therefore at $600\text{--}800^\circ\text{C}$ special constructions must be used with

3. Sputtering Ratios at 45 keV

Our sputtering results at 45 keV ion energy are reproduced in fig. 1. We used Cu, Ag and Ta as target materials because foils of high quality can be obtained commercially. The current density was $10\text{--}100\ \mu\text{A}/\text{cm}^2$ and the pressure in the target chamber was $0.8\text{--}1.5 \times 10^{-5}$ mm Hg. Usually we sputtered off more than $200\ \mu\text{g}/\text{cm}^2$, but in some cases lower amounts had to be accepted, since the sputtering ratio can be very low. It can also be

³⁾ K. O. Nielsen, Nucl. Instr. 1 (1957) 289.

⁴⁾ J. Druaux and R. Bernal, Proc. 1955 Harwell Isotope Conf., Ch. 4.

difficult to maintain an ion beam for a long time due to difficulties with the ion source or the charge of the material.

All targets for sputtering were handled carefully and cleaned with ether before first use. No attempts were made to smoothe the target surface. The ion current was always measured in the same Faraday cage and with an accuracy of ± 5 per cent. The micro-balance permits careful weighing within better than $\pm 5 \mu\text{g}$. This means an experimental accuracy of 7–10 per cent.

Some regularities in the sputtering ratio are to be noted:

3.1. SPUTTERING MAXIMA

Maxima of sputtering ratios occur for noble gas ions. The collected quantities are small, a few $\mu\text{g}/\text{cm}^2$. Therefore saturation is reached in a short time, and the sputtering ratio can be determined from eq. (2). Using an assumption made by Rol⁵⁾, it has been possible to present a sputtering formula for all noble gas ions of 5–65 keV bombarding different materials (see ref.¹⁾).

All three sputtering curves have about the same shape near the maximum points (noble gas ions). In addition, the general shape of the curves is repeated for each maximum. Thus the periodicity of the elements turns up also when it comes to sputtering. It is also to be noted that the sputtering ratio for related ions increases approximately proportional to the mass number. This is, however, a mere coincidence, since another ion energy would give a different mass dependence.

3.2. ZERO SPUTTERING

The final sputtering ratio is zero in a few cases. The reason is that the ions gradually build up a layer, which protects the underlying material. The condition for this must be that (i) the selfsputtering, that is the sputtering ratio of ions on the same target material, is less than one, (ii) the initial sputtering of the target material is not too large, (iii) the target temperature is sufficiently low to avoid evaporation of the deposit. Of course it takes a certain time for the ions to start building up a layer. At the beginning of the bombardment a high

⁵⁾ P. K. Rol, Thesis (Amsterdam 1960).

sputtering ratio would therefore be measured. After a while the conditions could be totally changed, and we should then measure an increase of weight instead of a loss. To study the process it is necessary to weigh the target plate at close intervals. We have thus interrupted the bombardment for weighing. If the weight of the target plate begins to increase, sputtering of backing material will soon be zero. According to eq. (3), where the in-

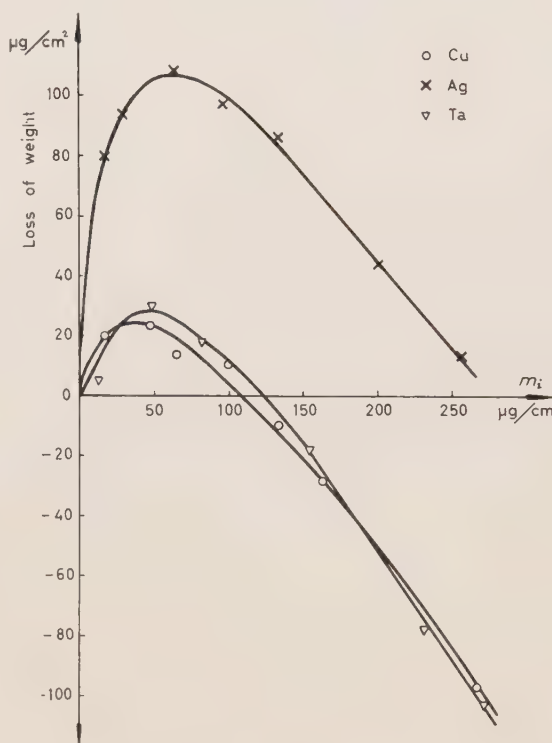


Fig. 2. Building-up of C ions.

coming amount is taken into account, we still measure a sputtering ratio, which is the selfsputtering of the material. Naturally the selfsputtering ratios measured in this way may be slightly different from those obtained by bombarding a rolled foil, since the deposit probably has a different structure.

To illustrate the process when S is zero, we give in fig. 2 the loss of weight of Cu, Ag and Ta targets as a function of the integrated amount of bombardment C ions. The ion beam was swept linearly over the target in order to obtain a uniform current density. A rectangular aperture of area 1 cm^2 placed immediately in front of each target plate cut off the

fringes of the beam. The current density was about $100 \mu\text{A}/\text{cm}^2$. No water-cooling of the target was used, which means a temperature of about 80°C at this current density. We see that after about $50 \mu\text{g}/\text{cm}^2$ of bombarding ions, all targets begin to increase their weights and the building-up corresponds to a selfsputtering of 0.4 atoms/ion. If the bombardment is continued for much longer a time the deposited carbon layer is likely to flake off because of the difference in thermal expansion of carbon and target material. Therefore one must be cautious, when very large amounts of bombarding ions are deposited.

Similar curves were obtained for Mg, Al, Si, Sc and Ti ions of 45 keV ion energy. For these ions an oxide formation on the target could eventually cause building-up because of a lower sputtering ratio of the oxide than of the pure metal. In order to determine whether the Mg target was oxidized in air at weighing, we made the experiment illustrated in fig. 2 both by weighing the same target plate at intervals and by weighing targets bombarded for different lengths of time. The results were the same. By letting in a controlled flow of dry oxygen we

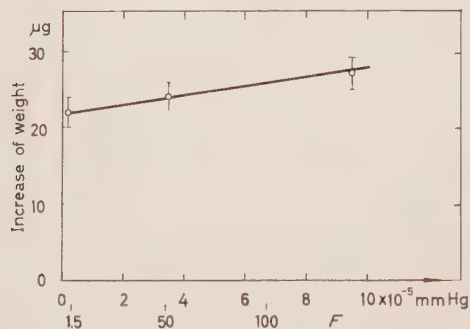


Fig. 3. Influence of pressure on Mg selfsputtering. F is the ratio between the numbers of O_2 molecules and Mg ions hitting the target.

have studied the selfsputtering ratio of Mg at different oxygen pressures in the target chamber. This investigation, shown in fig. 3, was made at a current density of $40 \mu\text{A}/\text{cm}^2$ with the target water-cooled. In fig. 3 the lowest ratio between the number of oxygen molecules and the number of Mg ions hitting the target was calculated from the assumption that 20 per cent of the lowest obtainable pressure was due to oxygen. However, an

unknown part of the pressure was due to gaseous products from the vacuum oil, although a liquid air trap placed near the target was used during the experiment. As seen in fig. 3 there is a small additional increase of weight because of the presence of oxygen, but it is unlikely that oxygen alone is the reason to building-up.

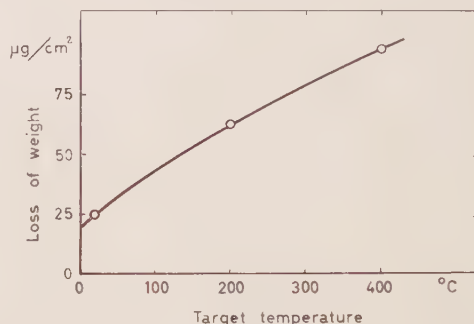


Fig. 4. Sputtered amount from Cu bombarded with Mg ions versus target temperature. Final sputtering ratio is not reached.

In fig. 4 the loss of weight of three Cu targets is illustrated for different target temperatures. All targets were bombarded with Mg ions at a current

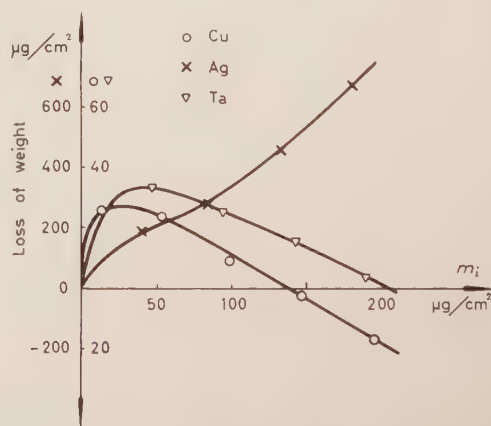


Fig. 5. 45 keV Ca ions bombarding Cu, Ag and Ta targets.

density of $50 \mu\text{A}/\text{cm}^2$ to $25 \mu\text{Ah}/\text{cm}^2$, which means that building-up had not yet started. We observe that the loss of weight increases strongly with temperature, indicating that building-up will not take place at sufficiently high temperatures. We have not investigated this further, but expect the temperature limit for building-up to be $150\text{--}200^\circ\text{C}$. If the current density is increased, the temperature of the outer layer of the target is also increased,

even at a good cooling of the target arrangement. This means that building-up will not occur at high current densities.

Ca ions build up on Cu and Ta but not on Ag. This is shown in fig. 5 for a current density of $60 \mu\text{A}/\text{cm}^2$ and the target water-cooled. For Ag the sputtering of the target material is too high to permit building-up. A final Ag sputtering of 1.5 is measured, which may be compared with the sputtering ratio 10.8 of equally heavy Ar ions on Ag. The dependence on target temperature and current density is expected to be similar to that found for Mg ions.

Building-up was also found for Be and B ions, but we did not make any complete investigation.

3.3. LOW SPUTTERING

V, Y and Zr ions demonstrate in an excellent way the process of saturation of a target material with bombarding ions. Figs. 6 and 7 show the measurements which we made at a low current density of

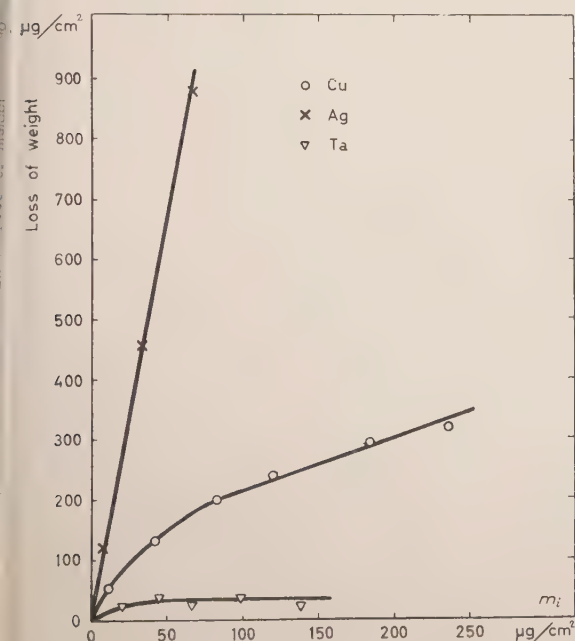


Fig. 6. 45 keV V ions bombarding Cu, Ag and Ta targets.

about $10 \mu\text{A}/\text{cm}^2$ because of the difficulty to get a suitable charge material for the ion source. However, we expect the same result up to at least $100 \mu\text{A}/\text{cm}^2$. Using an Ag backing the saturation of

ions is reached in a short time and the final sputtering is determined mainly by the properties of Ag, giving a fairly high final sputtering ratio. On Cu and Ta the built-in material changes the sputtering rate gradually. When saturation is reached, the loss of weight is proportional to the amount of bombarding ions, which means a constant final sputtering ratio.

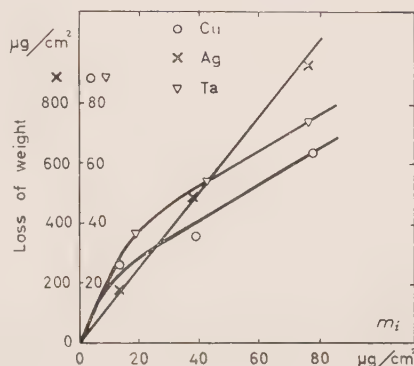


Fig. 7. 45 keV Y ions bombarding Cu, Ag and Ta targets. The curves for Zr are similar.

This ratio is thus representative for the obtained mixture and it is no longer determined by the backing material alone. There is no reason to give other sputtering ratios than those at saturation, since the ratio changes continuously from the initial value to the final ratio at saturation.

All the rare earth metals give results very similar to that of Y ions, and sputtering ratios have been determined at approximately the same experimental conditions for all of them, $10\text{--}20 \mu\text{A}/\text{cm}^2$. The sputtering ratios for the rare earths are of a magnitude expected from results with related elements discussed earlier.

When an investigation is made at a low current density and the sputtered quantities are small, it must be noted that different qualities of a target material can influence the result. In ref.¹), some phenomena are discussed, which give small deviations to sputtering results. On the other hand, much information would not be obtained if e.g. a large quantity were sputtered off directly.

For very light ions the sputtering is near zero. We have used He ions on Ag and measured a sputtering ratio below 0.1 atoms/ion. Li ions perhaps build up a layer, but we have not investigated this further.

3.4. INTERMEDIATE SPUTTERING

Most ions have intermediate sputtering ratios. The collection process is about the same as for noble gas ions. The only expected difference is that the diffusion rate for noble gas ions (ref.¹) is reduced to approximately zero for most other

Also the chemical binding between the collected particles and the target atoms can cause fluctuations in the curves. However, a systematic study of this fine structure has not yet been possible, since most of the fluctuations are of the same order of magnitude as the experimental error.

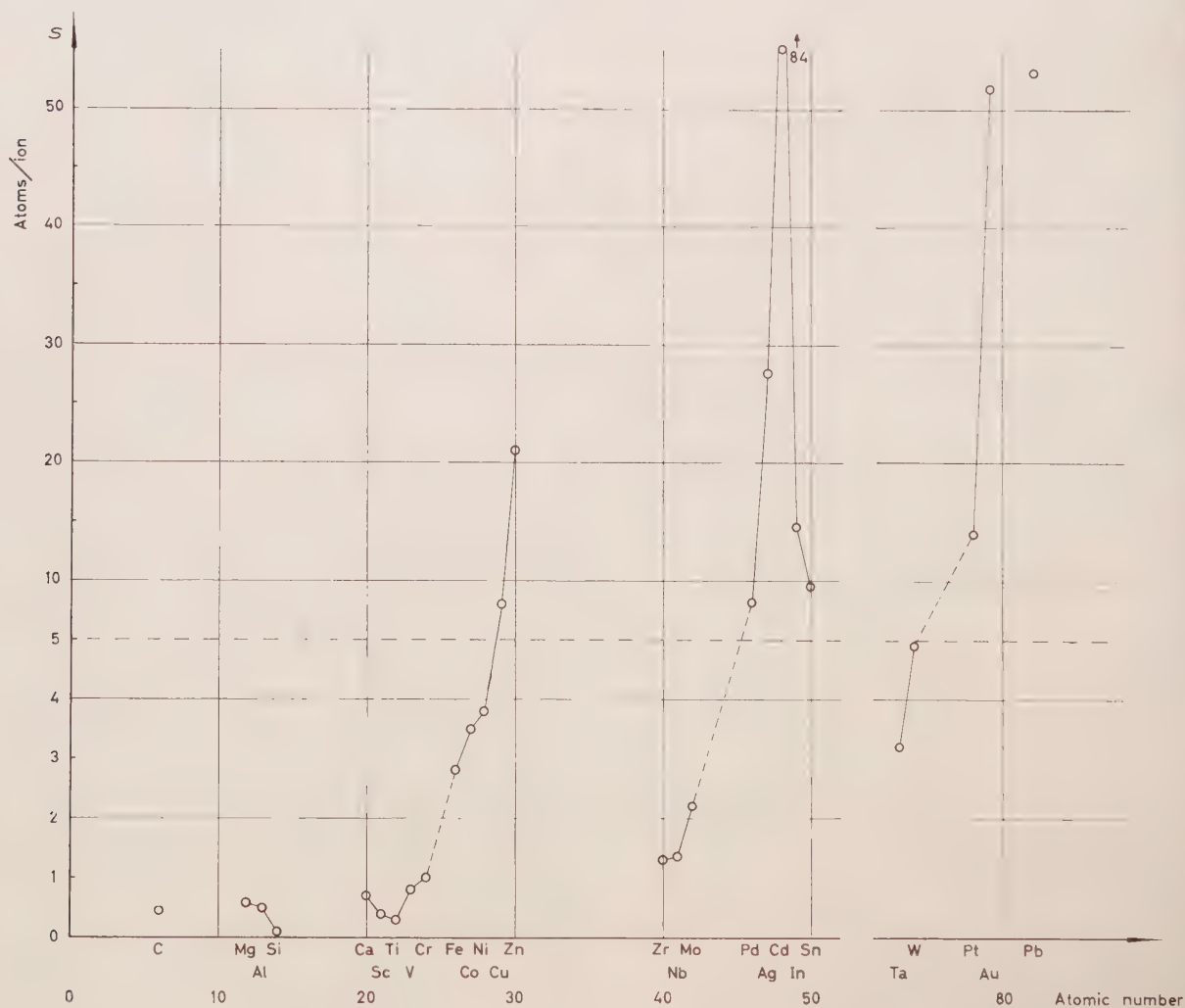


Fig. 8. Selfsputtering ratios for some materials at 45 keV ion energy.

elements. Diffusion might be of importance for such elements as Zn, Cd and Hg, but ions of these elements do not give extremely high sputtering ratios.

Where all three curves (fig. 1) show small, systematic variations of the sputtering ratio for an element, compared with its neighbours, (e.g. Ge-As-Se, Eu) it is in most cases due to a corresponding variation of binding energy of the ion elements.

4. Selfsputtering Ratios

In order to understand the physics of the sputtering process, it is important also to know the self-sputtering ratios. Fig. 8 shows the self-sputtering S_e for all elements, for which foils can be made without difficulty. S_e is calculated according to eq. (3) with $\eta_1 = 1$. In three cases, C, Si and Ca, the ratios were calculated from graphs, two of

which are shown in figs. 2 and 5. (Si similar to C.) Here also a periodicity is found in agreement with what could be expected from the periodic table. Compare fig. 18 of ref.¹). Maxima occur for Zn, Cd and Hg (extrapolated) and minima for the regions Sc-Ti, Y-Zr and the La series-Hf. The last is merely presumed, since metal foils have not yet been available. A comparison with binding energies of different elements indicates a relationship between the binding energy and the selfsputtering. According to Rol⁵) the energy dependence of sputtering ratios can be written

$$S = K \frac{E}{\lambda} \frac{M_1 M_2}{(M_1 + M_2)^2} \quad \text{atoms/ion} \quad (4)$$

where

E = energy of impinging particle;

M_1, M_2 = mass numbers of ion and target particle;

λ = mean free path of impinging particles

and, according to ref.¹) for noble gas sputtering

$$K = a \exp \left(-b E_B \frac{\sqrt{M_1}}{M_1 + M_2} \right) \quad (5)$$

a and b are constants;

E_B = binding energy of target atoms.

In the case of selfsputtering we have $M_1 = M_2 = M$, and eq. (5) should be written

$$K_e = a \exp \left(\frac{-b E_B}{2\sqrt{M}} \right). \quad (6)$$

In fig. 9 the experimental values of K_e are plotted in a logarithmic scale as a function of $E_B/\sqrt{M}$. For E_B we have used values tabulated in Thermodynamic Properties of Metals and Alloys⁶). Where data were not available in these tables we have used values given by Brewer⁷). As seen in fig. 9 nearly all the measured K_e can be represented by eq. (6). The exceptions will be discussed below. With numerical values of a and b we can write the final equation for selfsputtering

$$S_e \approx 4.24 \times 10^{-10} n_0 R^2 E \cdot \frac{1}{4} \exp \left(-11.2 E_B / 2\sqrt{M} \right) \text{ atoms/ion} \quad (7)$$

where

n_0 = target atoms per unit volume (in m^{-3});

R = collision diameter at ion energy E (in m);

E = ion energy (in eV);

E_B = binding energy (in eV).

This equation is identical with the equation for noble gas sputtering, given in ref.¹), except for the constant in the exponent. ($b = 10.4$ for noble gas sputtering.) The difference is probably due to a

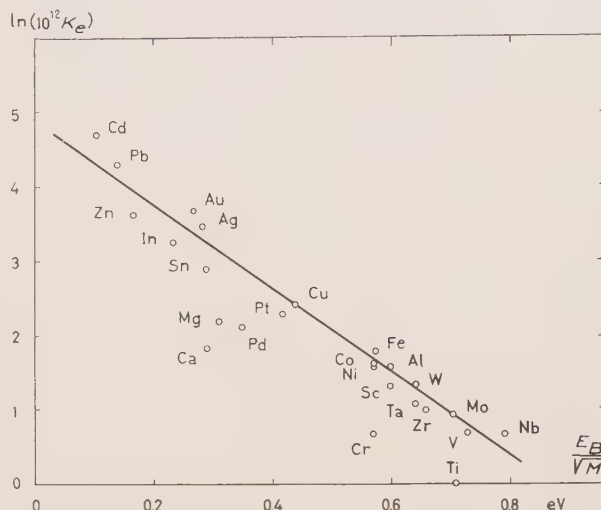


Fig. 9. $\ln K_e$ versus binding energy of the atoms divided by square root of the mass number.

reduction of the binding energy of the lattice, when noble gas ions are collected.

The most pronounced deviations from the systematics in fig. 9 were found for the elements which

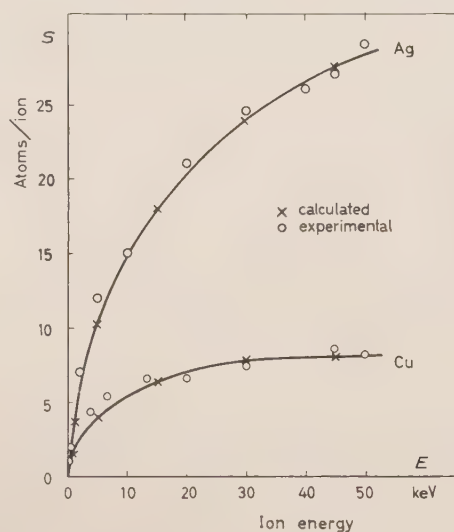


Fig. 10. Selfsputtering of Cu and Ag versus ion energy.

⁶) Thermodynamic Properties of Metals and Alloys, Minerals Research Laboratory, University of California, Berkeley.

⁷) L. Brewer, University of California Radiation Laboratory, Berkeley, UCRL 2854 (rev. 1955).

build up layers, since the layers have a different structure than the polycrystalline foils. A similar deviation was found for Cr sputtering on an electrolyzed deposit. Astonishing is the missfitting of Pd, for which we cannot find any explanation. This measurement has been checked carefully, giving the same result.

Of course errors in the tabulated binding energies can also cause deviations from eq. (7). However, for most elements the agreement is good, when considering both the experimental error of our measurements as well as that of the tabulated binding energies.

In order to establish if the selfsputtering ratio S_e also has the energy dependence given by eqs. (4) and (7), we have determined S_e for Cu and Ag as a function of the ion energy E (fig. 10). The fit is fairly good and eq. (7) can be used within the energy range investigated here.

5. Collected Quantities at 45 keV

In ref.¹⁾ we have experimentally determined the collected quantities in many backing materials of different noble gases after bombardment to saturation. This method to collect ions is called the direct collection method. We expect that other ions also can be collected up to a certain saturation value. In eq. (1) of ref.¹⁾ we have calculated the composition, f_s , of the material leaving the target because of sputtering

$$f_s = \frac{\eta_i - D}{\eta_i - D + S} \quad (8)$$

where η_i = initial collection efficiency;

D = fraction of impinging ions leaving because of diffusion at saturation;

S = sputtering ratio.

If we assume the mean concentration $\bar{f}$ of trapped atoms within the penetration depth, d_g , to be proportional to f_s , we get the collected quantity as

$$q_g \sim d_g \frac{M_g}{M_2} \frac{\eta_{ig} - D_g}{\eta_{ig} - D_g + S_g} \quad (9)$$

where g indicates that the equation is valid for a noble gas, with mass number M_g .

For ions of another element with mass number M_s , we can write analogously

$$q_s \sim d_s \frac{M_s}{M_2} \frac{\eta_{is} - D_s}{\eta_{is} - D_s + S_s} \quad (10)$$

In most cases $\eta_i - D < S$ and can be neglected compared to S . For small values of f , $\eta_i - D$ depends mainly on the properties of the target material. It should be noticed here that D_s takes into account both the diffusion and the sputtering of collected atoms. To get a rough estimate of q_s we chose the case $M_s = M_g$. Then we have $d_s \approx d_g$ and we assume that $\eta_{ig} - D_g \approx \eta_{is} - D_s$ and get for a target material

$$q_s \approx q_g \frac{S_g}{S_s} \quad (11)$$

To be able to calculate q_s for mass numbers between those of two noble gases, we assume that there exists a hypothetical noble gas for each mass number. The values q_g and S_g for this hypothetical gas can be obtained by interpolation between the experimental values. For a Ta target the values can be taken from figs. 11 and 1, respectively.

In fig. 11 q_s -values are also shown, calculated from eq. (11) for a Ta backing. For ions which give a low q_s , we can expect a good agreement with experiments, while the higher q_s -values are rather uncertain due to the approximations above. When $S_s = 0$, q_s can be infinite, since the ions build up a layer.

Experimental verification of the curve in fig. 11 is available in a few cases. From the sputtering curves in fig. 7, a saturation of Y ions in Ta can be estimated. Saturation is reached where the sputtering ratio becomes constant and this gives $q_s = 12 \mu\text{g}/\text{cm}^2$. A similar graph for Zr ions has given $15 \mu\text{g}/\text{cm}^2$. Using Rb and Yb ions on Ta, saturation values of 4.2 and 4.9, respectively, were determined by the tracer method. We have also collected Cr and Fe ions for different $\mu\text{Ah}/\text{cm}^2$ and compared the (p,γ) resonance curves obtained at a high voltage machine. These collection experiments have given 10 and $6 \mu\text{g}/\text{cm}^2$, respectively[†]. All

[†] After a bombardment of $20 \mu\text{Ah}/\text{cm}^2$ a maximum of about $20 \mu\text{g}/\text{cm}^2$ Cr was collected, and in the same way after $10 \mu\text{Ah}/\text{cm}^2$ about $10 \mu\text{g}/\text{cm}^2$ Fe. However, these amounts decreased after a prolonged bombardment to the values given above.

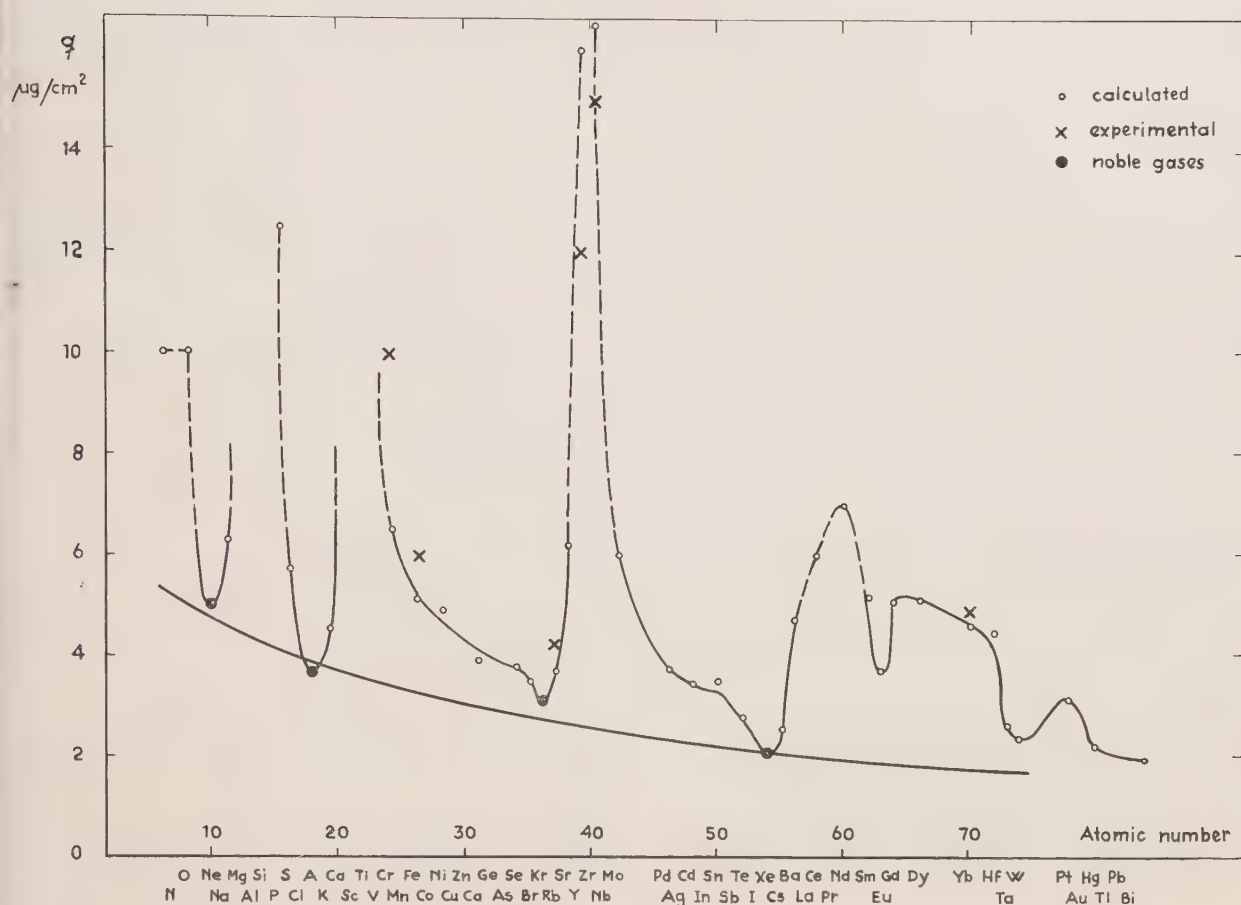


Fig. 11. Experimental and calculated saturation values for 45 keV ions bombarding a Ta target.

these experimental points (see fig. 11) are approximately in agreement with our simple equation for calculation of saturation values.

6. Direct Collection of Large Quantities

In § 3.2 the conditions for building-up are given. Of course the fundamental condition is that self-sputtering must be less than one, if large quantities are to be collected. However, if the ion energy is decreased sufficiently, we will reach the situation where $S_e = 1$, according to eq. (7). Below a certain energy it should thus be possible to collect unlimited quantities of any solid element by means of the direct collection method. In principle it is possible to retard the ion beam by having the collector at high tension. However, the retarding field acts as an electrostatic lens, which causes defocusing of the beam at low rest energies. By using

TABLE I
Ion energies for which $S_e = 1$, calculated from eq. (7). K_e values were taken from the straight line in fig. 9. Corresponding experimental sputtering ratios are also given. (No correction was made in eq. (7) for threshold energies¹¹.)

Element	E (keV)	S_e (experimental)
Fe	1.3	1.1
Ni	1.3	1.0
Cu	0.40	1.0
Zr	10.5	
Nb	25	
Mo	6.6	
Ag	0.20	1.0 (extrapolated)
Sn	0.45	1.0
Cd	< 0.05	
Ta	3.7	
W	3.0	
Pt	0.5	

¹¹) G. K. Wehner, Phys. Rev. 102 (1956) 690.

a suitably shaped collector arrangement it has been possible to overcome this defocusing⁸).

The problem is now to find the ion energy to which the ions must be retarded in order to start building-up. In § 4 we have found that eq. (7) for S_e fits our experiments fairly well down to energies of a few keV. If we assume eq. (7) to be valid also for a few hundred eV, this equation can be used to calculate the energy where $S_e = 1$. This was done for some ions and the result is given in table 1, together with a few experimental determinations of sputtering ratios at the calculated energy.

All the measured sputtering ratios are equal to 1 within the limits of experimental error ± 0.1 atoms/ion. Of course it is necessary to retard somewhat below the energies given in table 1 to get a reasonable collection efficiency.

7. Discussion of Sputtering Ratios

During the last years sputtering ratios have been determined by many authors, but no systematic studies have hitherto been made. Most of the results reported in literature are in agreement with our measurements. However, some details indicate that experimental conditions may influence the measurements a great deal. This is the case for some of the ions reported by us as building up layers. E.g. Rol⁵) has measured the sputtering ratio of Si ions bombarding on Cu and found it quite "normal" ($S \approx 4$ at 20 keV). As mentioned above we have found Si to build up at a low current density, $10 \mu\text{A}/\text{cm}^2$. Smith⁹) has also reported Si to build up, but no complete information on the experimental conditions is given. Guseva¹⁰) has measured the sputtering ratio of Si ions bombarding on Cu for different current densities. This author has found that the sputtering ratio increases approximately proportional to the current density below $200 \mu\text{A}/\text{cm}^2$ and is constant above 200.

There are many possible explanations of the discrepancies between the results:

a. formation of a resistant oxide layer protecting the Si deposit;

b. destruction of a Si deposit at high current density, because of the low thermal conductivity of Si;

c. sputtering ratio of Si-Cu mixture strongly dependent on target temperature.

In order to solve the problem definitely, it would be necessary to make sputtering experiments in very high vacuum (better than 10^{-8} mm Hg) with different current densities or target temperatures.

In the same way building-up of other ions at high ion energy is probably strongly dependent on experimental conditions such as current density, target temperature and composition of rest gas in the vacuum chamber. Particularly the oxygen content can be expected to influence the result. An interesting experiment in this connection is the building-up of C, which either indicates that the probability of oxide formation must be low, or that the oxygen pressure is lower than estimated from pressure measurements.

In all cases where very small current densities are used, there is also a possibility for a carbon layer to be formed from cracked vacuum oil, which can influence the sputtering result. In our separator freon-cooled baffles are used, and sometimes also a liquid air trap, placed near the target chamber. However, in spite of these arrangements influence from carbon formation cannot be excluded entirely.

For Mg and Ca, sputtering is influenced by the target temperature as mentioned above. Ti is reported by Smith⁹) to build up. However, the same author has also ascribed this property to Sn and W. According to our measurements both elements have a self-sputtering well above one. In order to study whether the reported discrepancies were due to different target temperatures, we bombarded heated Cu targets with Sn ions. No substantial influence of temperature could be detected below 500°C . Nevertheless other circumstances, hitherto unknown, may have caused building-up in the experiments reported by Smith.

Besides these building-up questions, there are many other sputtering problems, requiring further experiments to be made. To get complete and accurate information, it is of great importance to compare results from different experimental

⁸) B. Elbeck, M. C. Olesen and O. Skilbreid, Nucl. Phys. **10** (1959) 294.

⁹) M. L. Smith, Proc. 1955 Harwell Isotope Conf., Ch. 7.

¹⁰) M. I. Guseva, Fizika Tverdogo Tela **1**, N 10 (1959) 1540.

arrangements, and also to attack the problem along lines other than has been possible with the instrumentation available so far.

Acknowledgements

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Atomforskning, which we acknowledge with gratitude. We are much indebted to Prof. N. Ryde for placing working facilities at our disposal. We also wish to express our thanks to Ing. R. Timle for technical assistance and to the members of the Van de Graaff group at our laboratory for their (p,γ) resonance measurements.

PHOTOFRACTIONS FOR SODIUM IODIDE CRYSTALS

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Measurements of the pulse-height spectra obtained from large crystal NaI(Tl) scintillation spectrometers exposed to monoenergetic gamma rays have been used to determine the photofractions (ratio of total-absorption peak area to total spectrum area) for the crystals. The NaI crystals used were: (a) 4 inch diameter by 4 inch long; (b) 5 inch diameter by 6 inch long with a $\frac{3}{4}$ inch diameter by $1\frac{1}{2}$ inch deep well drilled in the end; (c) $9\frac{3}{8}$ inch diameter by $8\frac{5}{8}$ inch long with a $\frac{3}{4}$ inch diameter

by $2\frac{1}{2}$ inch deep well drilled in the end. The units were exposed to collimated monoenergetic gamma rays from 0.279 MeV to 2.76 MeV. The dependence of photofraction value on gamma-ray beam diameter (collimation) was determined for the 4 inch by 4 inch crystal. The photofraction values were fitted to an empirical relationship involving crystal radius and length, with three gamma-ray energy dependent parameters as the fitting variables.

1. Introduction

A useful parameter for evaluating scintillating crystals for gamma-ray spectrometers is the photofraction. This quantity is the ratio of the number of incident gamma-ray photons which lose all their energy in the crystal to the number which interact to any degree with the crystal. The photofraction can be experimentally determined from the scintillation spectrometer pulse-height spectrum due to monoenergetic gamma rays as the ratio of the number of pulses in the total-absorption peak (full-energy peak or photopeak) to the number of pulses in the entire spectrum. The photofraction is used in determining relative yields of several gamma-ray lines from a source whose spectrum has been measured with a crystal spectrometer. It is also used as an aid in selecting the most satisfactory crystal size for use in a particular experiment, and in constructing the pulse-height response function for the crystals to monoenergetic gamma rays. The value of the photofraction is a function of the size and shape of the scintillator, the energy of the gamma rays to which the crystal is exposed, and the source-crystal geometry.

A number of computational and experimental studies have been made of the response of NaI(Tl) crystals to gamma-ray photons. Most of these studies are referenced in the recent report by Davisson and Beach¹). Reference 1 describes a Monte Carlo calculation which gives the results of

photon interactions in sodium iodide scintillation crystals. Photofractions are computed and compared with earlier work. Calculations of this type have the advantage of being able to study a wide range of crystal sizes and source-crystal geometries, so that one can provide tables and curves of the dependence of photofraction on size and geometry parameters. Experimental results^{2,3}) have been compared^{4,5,6}) with theory for a limited number of gamma-ray energies and crystal sizes. The agreement is fairly good, although experimental values are generally somewhat lower than theoretical values. This is thought to be due to the presence of scattering materials, either around the source, in the crystal container or gamma-ray collimator or in associated materials.

This paper will describe the determination of the photofractions for several large NaI(Tl) crystal scintillation spectrometers exposed to monoenergetic gamma rays in a collimated beam geometry. This geometry provides the maximum photofraction values for a given crystal size, thus

¹) Charlotte M. Davisson and L. A. Beach, A Study of Photons in Sodium Iodide Scintillation Crystals, Naval Research Laboratory Report 5408 (December 31, 1959).

²) W. E. Kreger, Phys. Rev. **96** (1954) 1554.

³) R. S. Foote and H. W. Koch, Rev. Sci. Instr. **25** (1954), 746.

⁴) Martin, J. Berger and J. Doggett, J. Research Natl. Bur. Standards **56** (1956) 355 and Rev. Sci. Instr. **27** (1956) 269.

⁵) Maeder, Müller and Wintersteiger, Helv. Phys. Acta **27** (1954) 3.

⁶) Miller, Reynolds and Snow, Rev. Sci. Instr. **25** (1957) 717.

providing the most nearly unique response to gamma rays (minimum Compton-scatter distribution). This geometry is used to simplify the spectrum unfolding problem⁷) when several gamma-ray energies are involved, and also to permit determination of angular distributions of gamma radiation fields. Since the photofraction for a given crystal size and gamma-ray energy is dependent on collimator (and thereby photon beam) diameter, an experimental measurement was made of this dependence for one of the crystals. Comparison of the results with theory is made and the photofraction versus energy data is fitted to empirical curves based on crystal size and three parameters, one of which is adjusted to data and two of which are theoretically predicted⁴).

2. Description of the Experiment

Three different NaI crystals were used in this experiment. One was a Harshaw Chemical Company right cylinder 4 inch diameter by 4 inch long with a glass window at one end of the hermetically sealed package. The crystal unit was optically coupled to a 5 inch diameter Dumont photomultiplier. The second unit was a Levinthal Electronics scintillation transducer unit 6 inch long by 5 inch diameter with truncated conical end and a well $\frac{3}{4}$ inch diameter by $1\frac{1}{2}$ inch long drilled in the end. This crystal was directly coupled to a Dumont 5 inch diameter photomultiplier tube. The third unit was a Harshaw $9\frac{3}{8}$ inch diameter by $8\frac{5}{8}$ inch long NaI crystal with a truncated conical end and a $\frac{3}{4}$ inch diameter by $2\frac{1}{2}$ inch long well drilled in the end. This unit had four 3 inch diameter Dumont photomultiplier tubes coupled to four individual windows in the large end of the crystal. The crystal photomultiplier systems were mounted inside thick lead housings with provisions in one end of the housing for 8 inch long cylindrical collimators of different diameters for the incoming gamma rays. These systems provided for a beam of gamma rays of circular cross section incident on the front surface or well of the crystal and co-axial with the crystal axis.

⁷) N. E. Scofield, A Technique for Unfolding Gamma-Ray Scintillation Spectrometer Pulse-Height Distributions, U.S. Naval Radiological Defense Laboratory Technical Report 447 (September 1960).

The pulse-height distributions due to monoenergetic gamma rays incident on the crystal were recorded in a 100 channel pulse-height analyzer. The sources were placed on the collimator axis several inches away from the collimator. Sources were made up in a manner to minimize self-ab-

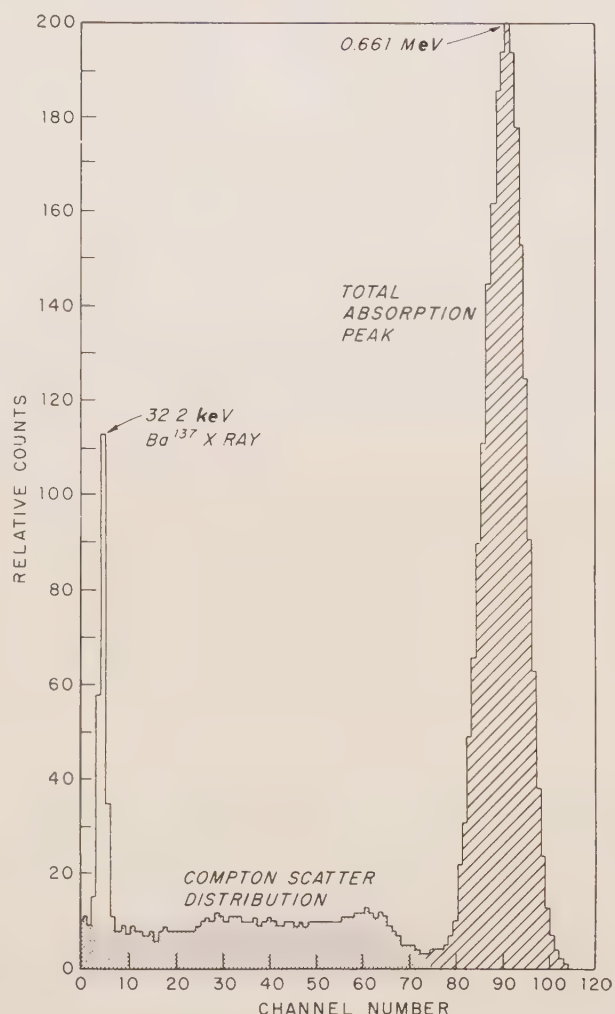


Fig. 1. Pulse-height spectrum for a 4 inch diameter by 4 inch long NaI crystal exposed to collimated 0.66-MeV gamma rays.

sorption and scattering in the source material. Background runs were made for each source spectrum by moving the source off the collimator axis so that no direct source gamma rays could reach the crystal. For the 4-inch crystal, measurements were made with collimators having hole diameters of $\frac{1}{2}$ inch, $\frac{3}{4}$ inch, $1\frac{1}{8}$ inch and $1\frac{1}{2}$ inch. The measurements using the other crystals used

TABLE I
Photofraction values

4" D. $\times$ 4" long NaI crystal

E_γ (MeV)	r/R						
	0	0.125	0.1875	0.2815	0.312	0.375	1.0
0.279	0.89*	0.88	0.87		0.87	0.86	0.81
0.411					0.85		0.67
0.51	0.84*	0.83	0.83		0.82	0.82	
0.66	0.75*	0.74	0.73		0.73	0.72	0.57
0.76					0.69		0.53
1.11	0.60*	0.58	0.57		0.57	0.55	0.44
1.28	0.54*	0.53	0.51	0.51	0.51		
1.85	0.53*	0.51	0.51	0.49	0.49		
2.76					0.48		

* linear extrapolation from data.

5" D. $\times$ 6" long NaI with $\frac{3}{4}$ " D. $\times$ $1\frac{1}{2}$ " deep well

E_γ (MeV)	$r/R = 0.1$
0.279	0.93
0.32	0.94
0.48	0.91
0.51	0.89
0.66	0.82
0.76	0.79
1.11	0.67
1.28	0.63
1.85	0.56
2.76	0.52

$9\frac{3}{8}$ " D. $\times$ $8\frac{5}{8}$ " long NaI with $\frac{3}{4}$ " D. $\times$ $2\frac{1}{2}$ " long well

E_γ (MeV)	$r/R = 0.053$
0.41	0.945
0.48	0.92
0.66	0.86
1.11	0.75
1.28	0.72
1.85	0.64

$\frac{1}{2}$ inch diameter collimators. Figure 1 shows a typical pulse-height spectrum (with background subtracted) for the 4-inch crystal using monoenergetic gamma rays from Cs¹³⁷. Significant features of the spectrum are the total-absorption peak (often called photopeak), and the Compton-scatter distribution due to interactions by gamma rays in the crystal where less than the total photon energy is transferred to the crystal.

3. Results

The ratio of the total-absorption peak area to total spectrum area (photofraction) was calculated from the pulse-height distributions for each of the conditions of incident gamma-ray energy and collimation. The values obtained for the various crystals are shown in table 1 as a function of

gamma-ray energy and collimator radius, r . In the case of sources emitting two gamma rays, it was necessary to subtract out the effect of the pulse-height distribution from the low-energy gamma ray in order to calculate the photofraction for the high-energy gamma ray. The values shown for the 4 inch by 4 inch crystal with $r = R = 2$ inches are from old data and may not be as reliable as the rest of the data.

Since a large variety of collimator sizes are used by different experimenters, a portion of the data were examined to determine the effect of collimator size on photofraction. A convenient parameter for describing collimation for cylindrical collimators is the ratio of the collimator radius, r , to the crystal radius, R . Figure 2 shows peak to total ratios (photofractions) as a function of r/R for six different

incident gamma-ray energies on the 4 inch by 4 inch NaI crystal. In the notation of Berger and Doggett⁴) the photofraction values for $r/R \neq 0$ are called p_{bb} or broad-beam photofractions. We have extrapolated the data back to $r/R = 0$ using a

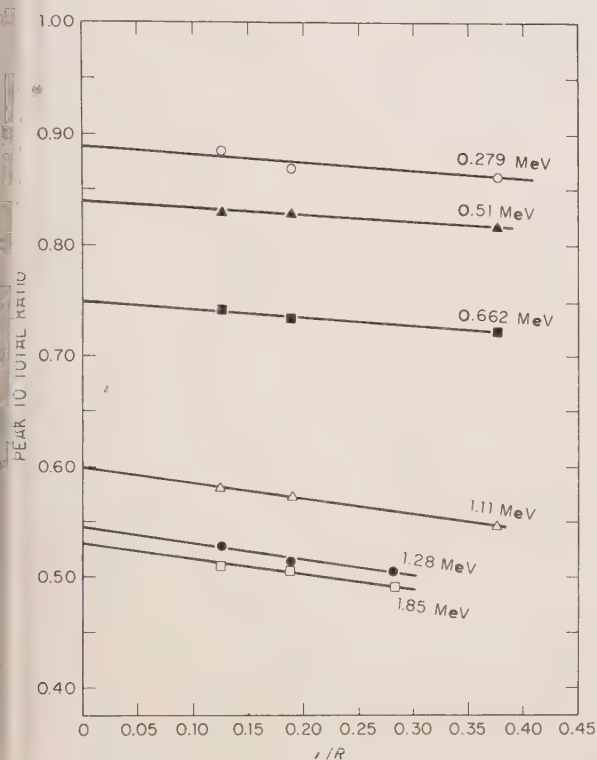


Fig. 2. Peak area to total spectrum area (photofractions) versus the ratio of collimator radius r , to crystal radius R for a 4 inch diameter by 4 inch long NaI crystal exposed to the indicated monoenergetic gamma rays.

linear extrapolation and have obtained the p or narrow beam values which are similar to the case for which Berger and Doggett have calculated values. Using these extrapolated values we have calculated the ratio of the broad beam p_{bb} to the narrow beam p and fig. 3 shows a plot of this ratio for the different energy gamma rays and different r/R values. The solid lines fit the experimental points of the ratio for a 4 inch by 4 inch crystal, while the dashed line is a curve calculated by Berger and Doggett for 0.66 MeV gamma radiation and a 5 inch diameter by 4 inch long NaI crystal. Although their crystal size is slightly different from ours, this comparison is reasonable, since Berger and Doggett⁴) show (their fig. 2) that the

p_{bb}/p vs r/R curves are not very different for different crystal sizes at a given gamma-ray energy. The calculation by the Monte Carlo method therefore predicts a more serious broad-beam effect than we see experimentally.

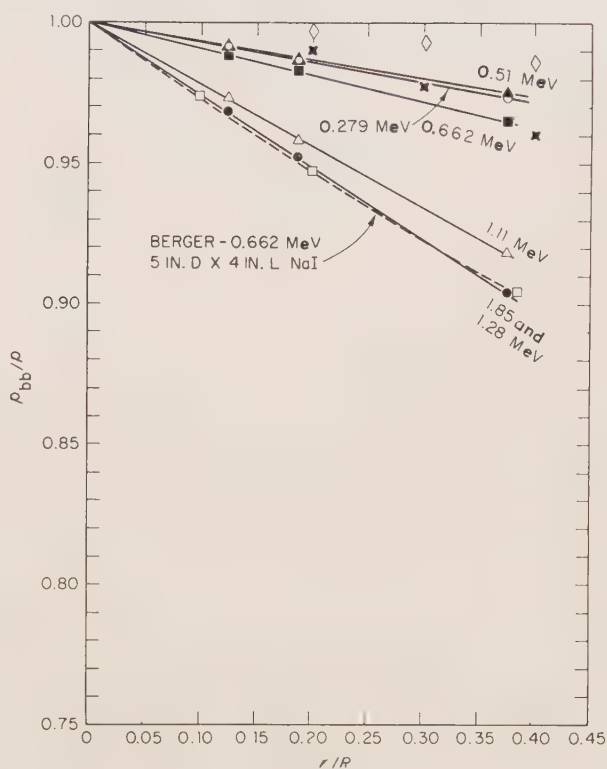


Fig. 3. Broad beam photofractions divided by narrow beam photofractions versus r/R . 4 inch diameter by 4 inch long NaI crystal exposed to indicated gamma-ray energies. Dashed curve from ref.⁵) ◇ diamond points and × points using method of ref.⁴).

If one follows the work of Maeder *et al.*⁵), a slightly different treatment is used. An expression is developed to predict the values of p_{bb} between the limiting cases $r \rightarrow 0$ and $r \rightarrow R$. Converting their notation to that of Berger and Doggett⁴), this expression is:

$$\frac{p_{bb}}{p} = 1 - \left(\frac{r}{R}\right)^2 \left[1 - \frac{P_{bb}(r=R)}{p}\right]. \quad (1)$$

Substituting the extrapolated p values (linear extrapolation) of fig. 2, and the old experimental results for p_{bb} for $r = R = 2$ inches, we find that the bracket term in formula (1) is 0.09 at 0.279 MeV and about 0.26 at 0.51, 0.66 and 1.11 MeV. This gives the points indicated as ◇ on the plot in

fig. 3 for 0.279 MeV and $\times$ for the three higher energies. Although ref.⁵⁾ treats the collimation correction further, it is not possible to apply their results here due to the fact that all the crystal sizes used in that work are smaller than those used in this work. It is noticed in the figure that treatment by the method of ref.⁵⁾ gives quite conservative values for p_{bb}/p compared with the actual ex-

the trend of the data and because we feel that at $E_\gamma = 0$, there should be no broadbeam effect. Although Berger and Doggett⁴⁾ have calculated p_{bb}/p for 0.662 MeV only, they estimated that within an error of only a few percent, the same fractional correction could be applied at other energies. This is true at small r/R values where the correction only amounts to a few percent, but it does not appear to

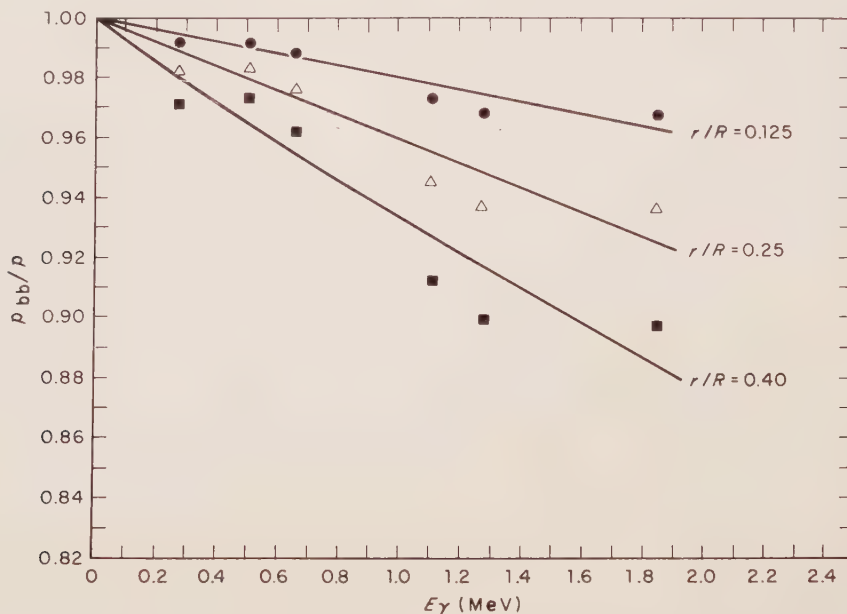


Fig. 4. p_{bb}/p ratios versus gamma-ray energy for selected r/R . 4 inch diameter by 4 inch long NaI crystal. Straight lines used only to indicate the trend of the data.

perimental values. However, our treatment following the method of Maeder *et al.*, has used p values extrapolated from experimental values by a linear extrapolation and $p_{bb}(r = R)$ values based on data of questionable reliability. Nevertheless, it would seem to indicate that the functional dependence given by formula (1) is not proper for this crystal size.

We would expect that the broad-beam effect should get worse as the energy of the gamma radiation increases. If the data of fig. 3 is replotted to show the energy dependence, it can be seen in fig. 4 that the ratios p_{bb}/p do depart more from 1.00 as energy increases. The data are not particularly consistent, however, which may indicate the degree of unreliability of the values. We have used straight lines going through zero in the plot only to indicate

be true for $r/R \leq 0.25$. Here, there is a marked energy dependence of p_{bb}/p in the experimental results.

We have followed the lead of Maeder *et al.*⁵⁾, and Berger and Doggett⁴⁾ in relating photofraction to $\sqrt{LR}$ by several constants. Here L is crystal length in cm and R , crystal radius in cm. Figure 5 shows a plot of the constants of the relationship given in the Berger article. The formula is also included. The parameter A is one minus the number albedo for gamma rays. The parameter B is related to the ratio of photoelectric absorption coefficient to the total-absorption coefficient. The parameter C is an adjustable parameter. The values of C plotted (circles and solid curve) on fig. 5 were solved from the formula on fig. 5 using the p values of table 1 for the 4 inch by 4 inch crystal and Berger and Doggett's⁴⁾ values of A and B . The dashed curve

for C presents the results obtained by Davisson and (each¹) using a Monte Carlo calculation of p and Berger and Doggett's values of A and B . It is apparent that a relatively good fit is obtained at values of $E_\gamma > 0.4$ MeV. The discrepancy below 0.4 MeV is probably in the experimental data. The point at $E_\gamma = 0.279$ MeV is taken with a Hg^{203} source, which is difficult to prepare in a thin scatter-free manner. It is likely that the predicted value, $p = 0.963$ for $E_\gamma = 0.279$ MeV using the C value of (ref.¹) more nearly represents the response to monoenergetic gamma rays at that energy.

Figure 6 displays some of the experimental photofraction values of table 1 for the crystals shown on the bottom of the figure. The lower curve gives the p_{bb} ($r = 1\frac{1}{2}$ inches) vs E_γ plot for the 4 inch by 4 inch crystal. The dashed curve above it gives the p ($r = 0$) vs E_γ plot for the same crystal. The next higher curve (circles) gives p_{bb} ($r = \frac{1}{2}$ inch) vs E_γ for the 5 inch by 6 inch crystal and the top curve shows p_{bb} ($r = \frac{1}{2}$ inch) vs E_γ for the 9 inch by 8 inch crystal. Since all the results shown were taken using the same gamma-ray sources and essentially the same source-collimator geometry, source self-absorption and scatter and collimator scatter should be about the same for all crystals.

We have used the formula of fig. 5 to predict the photofraction (p) values for the 5 inch diameter crystal and the $9\frac{3}{8}$ inch diameter crystal. Since the gamma rays are collimated into the crystal well and therefore most of the backscattered photons are absorbed in the sides of the well, the value of A becomes one for all E_γ . The calculation used the B values of Berger and Doggett as shown in fig. 5 and the C values calculated using the 4-inch crystal data, also as shown in fig. 5. In the case of 5 inch diameter by 6 inch crystal, $R = 2\frac{1}{2}$ inch and $L = 4\frac{1}{2}$ inch, the length of crystal from the well bottom to the back surface. The values of p obtained are shown as the solid circles in fig. 6. The predicted values agree quite well with the experimental values of p_{bb} for the crystal except at the high energy end. If one corrected the p_{bb} values to get the p values, an even better fit would be obtained. Using the p_{bb}/p values obtained with the 4-inch crystal to correct the p_{bb} values for the larger crystal, the correction would amount to around 2.5 percent at

E_γ around 2.0 MeV and about 1 percent at E_γ around 0.6 MeV, since $r/R = 0.1$. We did not make the correction since we feel the 4-inch crystal p_{bb}/p values may not apply for the larger crystal.

A similar calculation for the larger crystal used $R = 4\frac{1}{8}$ inch and $L = 6\frac{1}{8}$ inch and the same values of A , B and C to calculate p . The data of this calculation are shown in fig. 6 as solid triangles. It

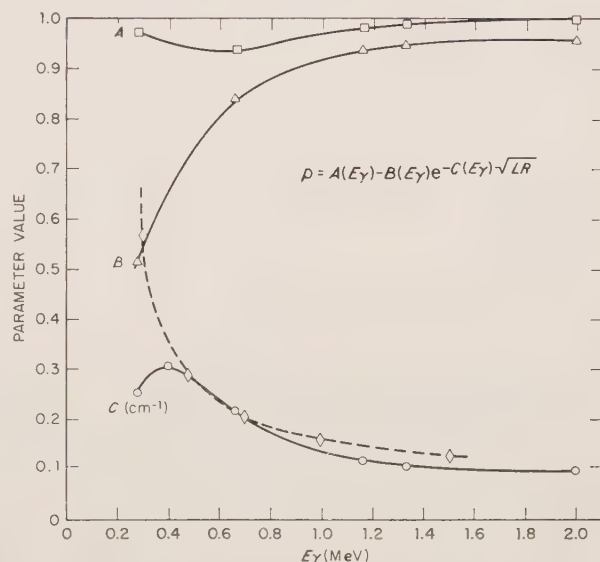


Fig. 5. Parameters for empirical photofraction formula. Values for A and B taken from ref.⁴). Solid curve for C values calculated from experimental data for 4 inch diameter by 4 inch long NaI crystal. Dashed line taken from ref.¹) (Monte Carlo calculation).

can be seen that the agreement is poor. Again, the correction to p values would be relatively small since $r/R = 0.053$, so that it is reasonable to compare the p values of the calculation with the p_{bb} values of the experiment. It appears from this result that it is not reasonable to use the constants obtained at a $\sqrt{LR}$ value of 7.17 cm (the 4 inch by 4 inch crystal) for a crystal having as large a $\sqrt{LR}$ value as 13.5 cm, as does the $9\frac{3}{8}$ inch diameter crystal. The 5 inch by 6 inch crystal has $\sqrt{LR} = 8.51$ cm and at this value the constants appear to be useful for predicting narrow beam photofraction values.

The errors in the experimental determination of the photofractions are difficult to evaluate. Data were taken so as to provide well over 10000 counts in the photopeaks, so that the statistical errors in

the calculation amounted to less than 1 percent. There was some question about the determination of the lower edge of the photopeak and this was made relatively unambiguous by reflecting the upper edge about the axis of symmetry of the photopeak. Variation of the calculated photo-

seem to agree with that suggested in ref.⁵). The narrow beam photofraction calculation parameter C predicted by theory⁴) agrees quite well with experiment, except at low values of E_γ where the experimental results may be questionable due to source difficulties. The values for parameter C in the

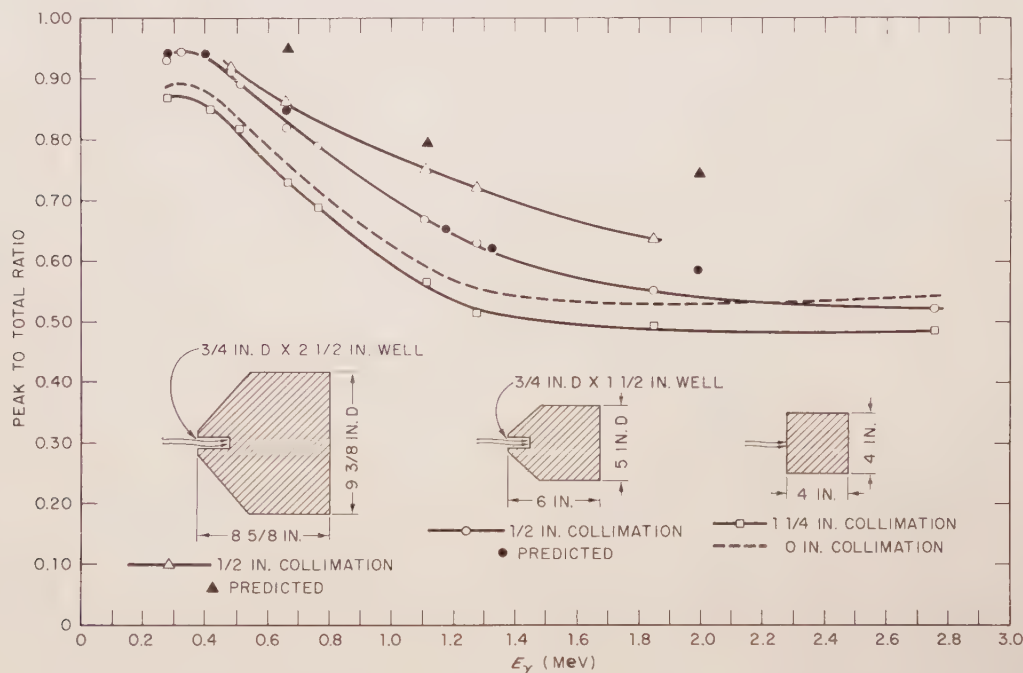


Fig. 6. Photofractions *vs* gamma-ray energy for crystals shown. "Predicted" values obtained using the formula and parameters of fig. 5 and proper values of crystal length L and crystal radius R .

fraction over several experimental runs with the same source amounted to ± 2 percent and the actual data in table 1 represents the average of 2 to 4 runs in most every case.

4. Summary

The correction of broad-beam photofraction values to narrow beam values has been determined experimentally for a 4 inch by 4 inch NaI crystal. The ratio of p_{bb}/p is found to be dependent on gamma-ray energy and collimator radius. The functional dependence of p_{bb}/p on r/R does not

expression (fig. 5) relating photofraction to $\sqrt{LR}$ seems to give p values comparable to experiment for a reasonable range of crystal sizes, as shown in ref.¹). Experimental results for crystals having $\sqrt{LR} = 7.17$ cm and 8.51 cm are shown to be in fairly good agreement with theory. However, the large crystal having $\sqrt{LR} = 13.5$ cm had p values considerably below those theoretically predicted by the same formula and theoretical parameters as with the smaller crystal. No conclusion is reached as to why the parameter C should be dependent on crystal size.

APPARATUS FOR AUTOMATIC TREATMENT OF PHOTOGRAPHS OBTAINED FROM BUBBLE CHAMBERS[†]

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Joint Institute for Nuclear Research, Dubna

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The present paper is a description of an automatic apparatus assigned to coordinate method application to photographs taken from bubble chambers. The accuracy of the coordinate measurements on the film is 2-4 microns. Tracking velocity

(over the film) reaches 3 mm/sec. The coordinate readings are punched on perforated tape, and they are ready to be put into electronic computers for further mathematical treatment.

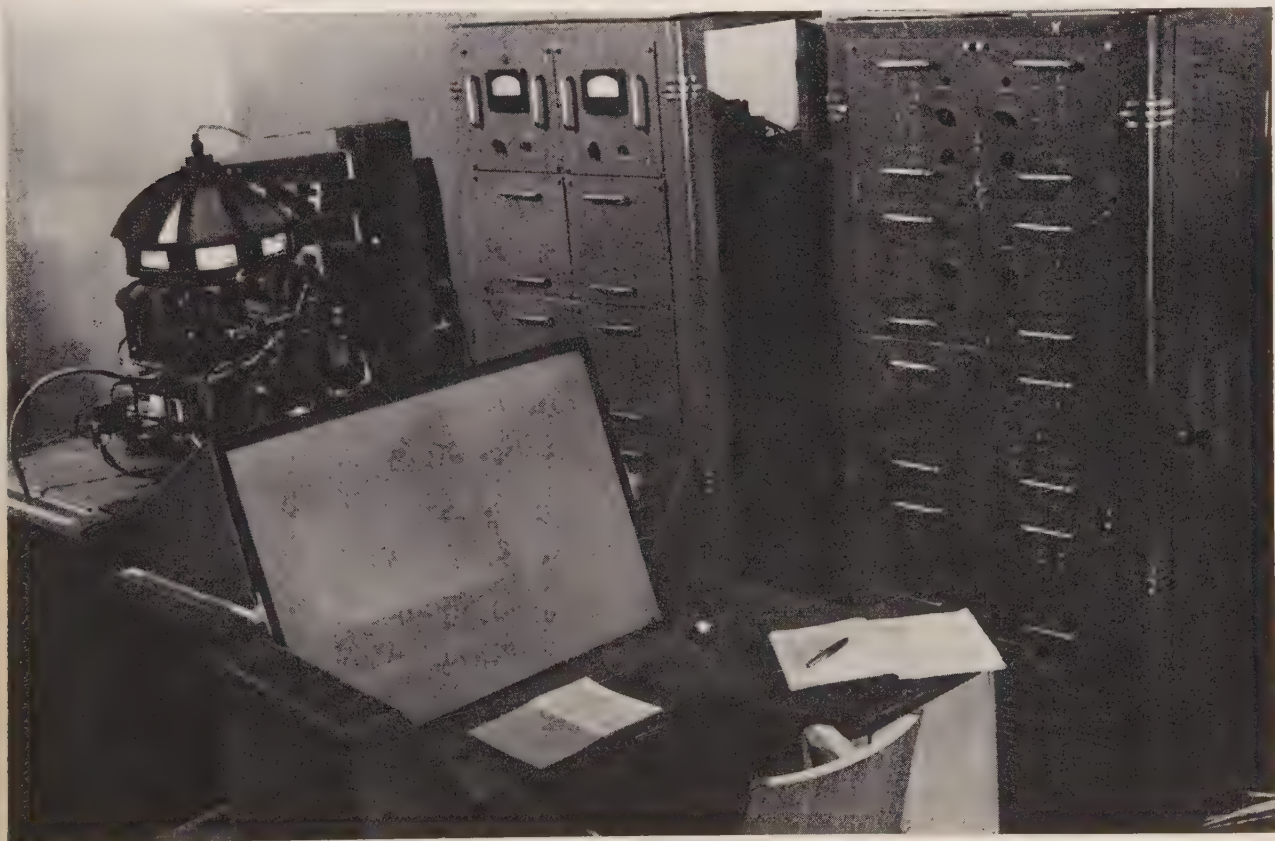


Fig. 1. General view of the apparatus. The automatic section is shown without the housing.

[†] The main features of the apparatus have been reported at the International Conference on High-Energy Accelerators and

Instruments in Geneva, 1959. (Proc. of the Int. Conf. on High-Energy Accel. and Instr. CERN, 1959).

1. Introduction

The bubble chamber technique has been widely used for several years in high-energy particle interaction research with high power accelerators. However, it is rather difficult to determine the space and energy characteristics of the particles from the results obtained in bubble chambers. The treatment of experimental data can be advanced by high speed electronic computers, the time required for coordinate measurements being reduced by automation of this process¹). The described apparatus for automatic photograph treatment "APT" is assigned for automatic coordinate reading and recording, the whole problem thus partly being solved. Each of the photographs of a stereoscopic pair is examined separately, following one after another, the real space image of the event being established in the computer.

2. General Characteristic of the "APT"

A general view of the apparatus (without the housing) and its operating principles are presented in fig. 1 and 2. The film 1 to be examined is placed on a measuring stage 2 which can be moved by two servo motors 3 and 4 in two perpendicular directions. The image of the photograph magnified 10 times is projected on a ground glass screen 8 (60 × 85 cm) placed before the operator. The projecting device consists of an illuminator 5, a lens 6 and a mirror 7, the middle part of which is semi-transparent. By means of a set of prisms 9 part of the image is directed through the semi-transparent section of the mirror to a photoelectric sensing 10.

The signals from the photoelectric sensing are used in the tracking unit to control the two servo motors which move the measuring stage, and a motor 11 which turns the photoelectric sensing itself. The tracking unit secures a proper succession of all the track points as they are projected on the photoelectric sensing, due to corresponding displacement of the measuring stage. Whenever a point of the track is projected on the center of the photoelectric

sensing, the coordinates of the measuring stage at the moment coincide with the coordinates of that point. To enable the operator to see which part of the track is being examined at the moment, a special device connected with the photoelectric sensing by

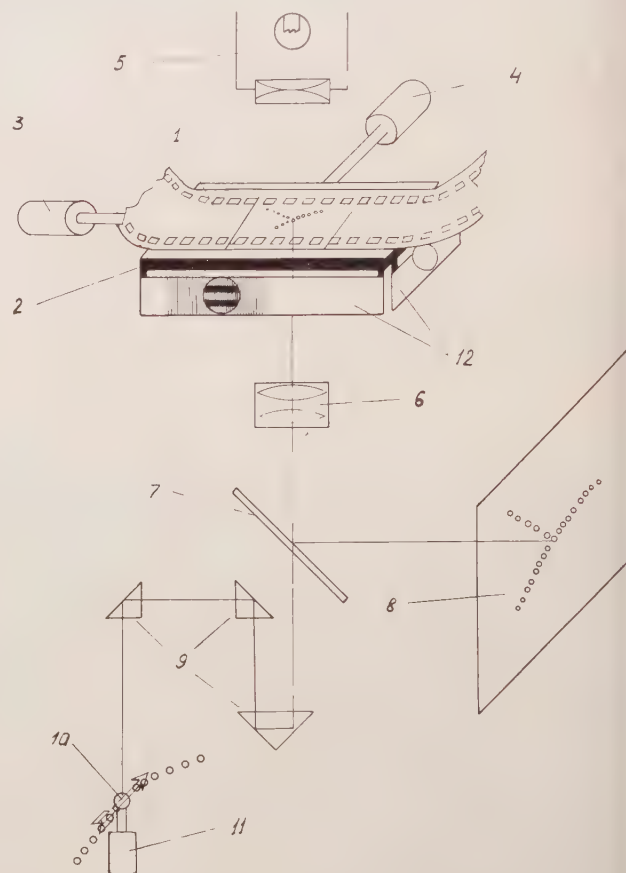


Fig. 2. Construction of the apparatus: 1. film; 2. measuring stage; 3 and 4. servo motors; 5. illuminator; 6. lens; 7. mirror; 8. screen; 9. prisms; 10. photoelectric sensing; 11. motor of the photoelectric sensing.

selsyn transmission projects an arrow shaped light beam on the screen. The arrow shows in which direction the movement is (or will be) proceeding, the rotation center of the arrow coinciding with that point of the track, the coordinates of which are being measured.

Linear displacement of the measuring stage is detected by means of diffraction gratings. The use of an intermediate memory circuit in the recording device permits the recording of the coordinates without stopping the whole system. Using the

¹) Proc. of the Int. Meeting on Instr. for the Evaluation of Photographs (Geneva, 1958);

Y. Goldschmidt-Clermont *et al.*, Nucl. Instr. 2 (1958) 146;

Proc. of the Intern. Conf. on High-Energy Accelerators and Instrumentation CERN, 1959.

binary code, the coordinates are punched on a perforated tape, and they are available to be fed into any of the computers: "Ural-I", "Ural-2" or "Kiev".

The main characteristics of the "APT":

1. Size of the frame – up to 60×120 mm.
2. Width of the film to be examined – from 35 to 80 mm.
3. Linear tracking velocity over the film – up to 3 mm/sec.
4. Accuracy of tracking a track of average density – ± 2 microns.
5. Division unit of the recording system – 2.5 microns (on the film).
6. Capacity of the coordinate counters – 16 binary divisions.

The stage from the instrument microscope "BMI" is used in the apparatus as a measuring stage. The image of the photograph is projected by a 105 mm focus "Tessar" photographic lens. In the illuminator an elliptic reflector is used giving a light concentration of about 50%. The illumination achieved on the screen (up to about 50 lux) is sufficient for work in a non-darkened room with a 100 W incandescent lamp ("SC" 62 V) with unsaturated filament current. The use of a low-power lamp in the apparatus makes cooling of the film and the illuminator superfluous. To minimize the heat flow through the film a heat filter is arranged – a 2 cm thick layer of 2.5% water solution of cuprous sulfate.

The film transport mechanism is mounted on the microscope stage. Construction of the transport mechanism allows three films to be placed on the stage simultaneously. It is possible to change the frames quickly on each side of one film as well as to switch to any of the three films being examined. The films are pressed to a flat parallel glass plate by vacuum suction. The film transport and switching from one film to another is directed from the control panel.

3. Tracking Unit

The tracking unit provides an accurate adjustment of the photoelectric sensing to the track and secures the tracking along it. The block diagram of

the tracking unit is shown in fig. 3. The photoelectric sensing has four photosensitive areas. 0.5×2.5 mm each. Position of the track relatively to these areas during the tracking process is shown in fig. 3. The image of the track is split into four parts by means of an optical set of rectangular prisms (fig. 4). At first the image is split into two, the separating line (in the center) being directed along the track. Then each of the two light beams is split, on its turn, into two again, and brought to the photocathode of a photomultiplier tube (type "FEU-35"). Photocurrents from the phototubes are compared in pairs as shown in the diagram (fig. 3). It can be seen from there that each pair of the phototubes (1 and 2, 3 and 4) is equally illuminated in the case if the middle part of the track coincides with the central line of the photoelectric sensing. Any deviation from the central line disturbs the illumination balance of the phototubes. The output signals of the comparison circuits are added to each other and subtracted from each other. The sum and the difference, thus obtained, are signals of parallel and angular incongruity, respectively, between the track and the central line of the photoelectric sensing. The angular incongruity signal after amplifying is transmitted to the motor which turns the whole photosensitive device so as to decrease the incongruity. Thus, the central line of the photoelectric sensing always tends to be directed approximately along the tangent of the track.

Direction of the measuring stage velocity vector is determined by a sine-cosine rotating transformer "SCRT-I" (fig. 3), the rotor of which rotates together with the photoelectric sensing. Tracking velocity is settled by the operator by means of changing the voltage applied to the exciting coil of the transformer "SCRT-I", the voltage being decomposed by the transformer into "X" and "Y" compounds, which after amplifying are applied to the corresponding servo motors.

The accuracy of the measurements is secured by parallel displacement control. The error signal, arising in a case of parallel displacement of the track relative to the photoelectric sensing, is decomposed by means of a sine-cosine rotating transformer "SCRT-2" into components parallel to the coordinate axes; these components are applied to the

corresponding servo motors so that the surplus velocity vector is perpendicular to the track.

The circuit of the input is shown in fig. 5. The electron flux in phototubes is modulated with frequency 400 c/sec, the modulating voltage about 5 V

Since the phototube output signals already have a 400 c/sec frequency, the rather simple alternating current circuits can be used for adding, subtracting and amplifying of them, and multiple transformations of the signals are avoided.

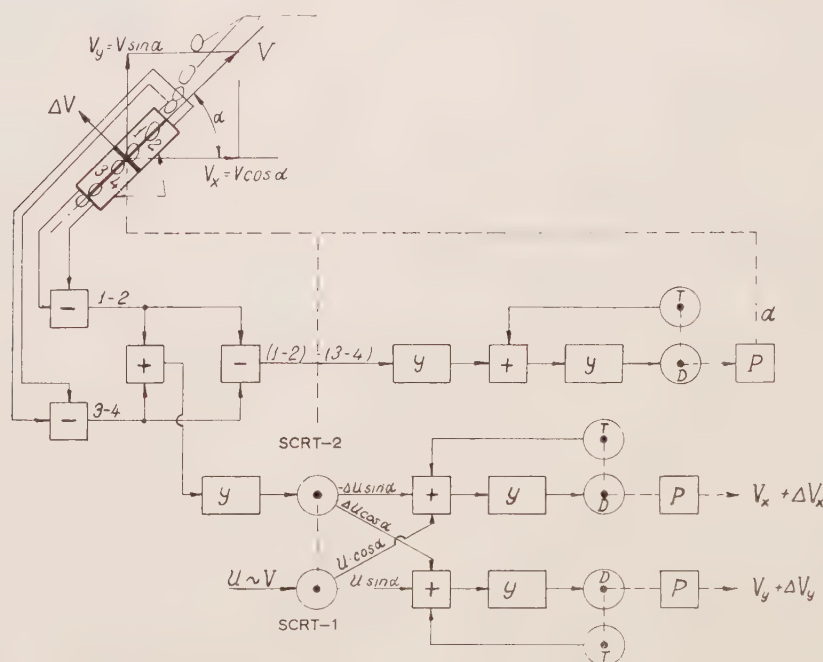


Fig. 3. Block diagram of the tracking unit: - - comparison (subtracting) circuits; + - adding circuits, y - amplifier, D - motor; T - speed voltage generator, P - gear.

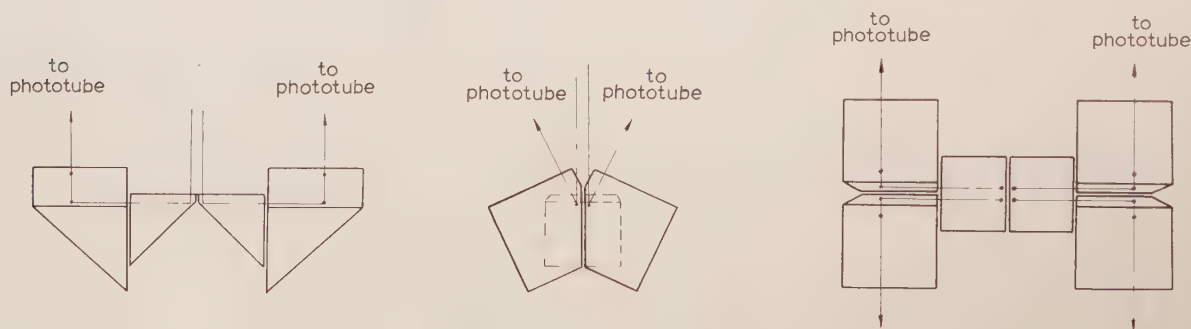


Fig. 4. Prism system of the photoelectric sensing.

being applied to the anode. The measurements have shown that the region of linear dependences of the phototube (type "FEU-35") output current on the voltage of the last dynode-anode interspace lies within the limits from 0 to ≈ 30 V. The operating point has been chosen at a level of about 15 V.

The use of a "static" type photosensitive device has certain advantages of a rather simple construction and simple design of the following electronic circuits; at the same time, it requires the selected phototubes to have similar characteristics. It is possible to keep the signal constant within the

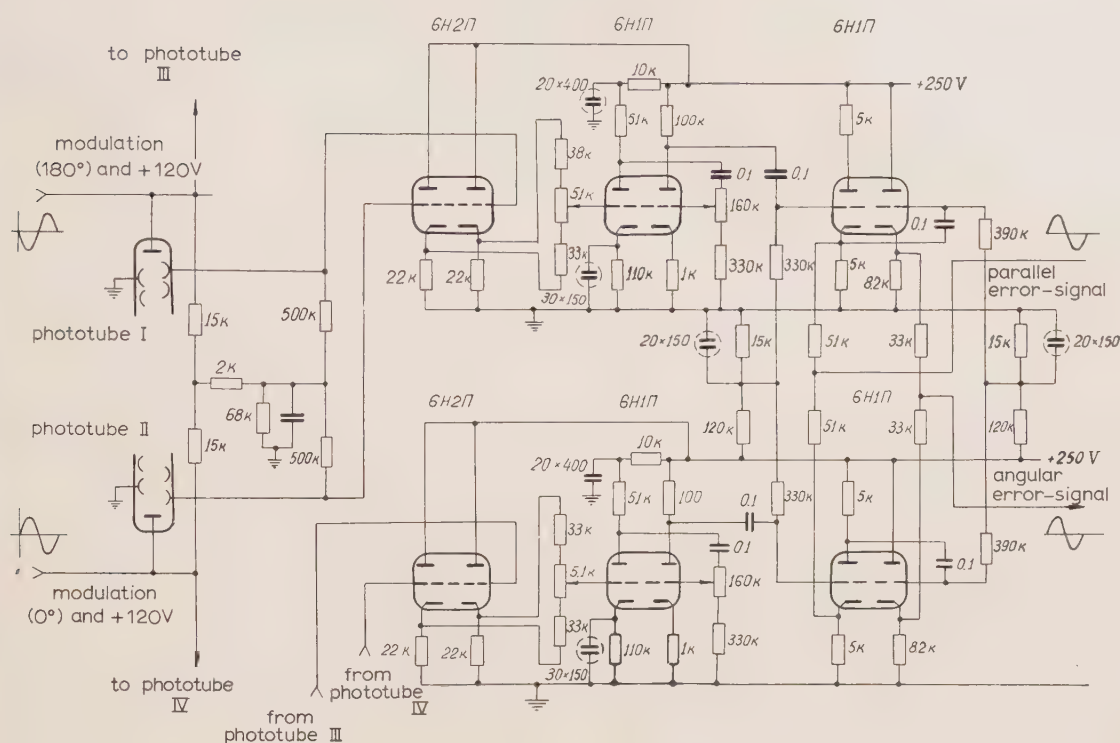


Fig. 5. Input circuit of the tracking unit.

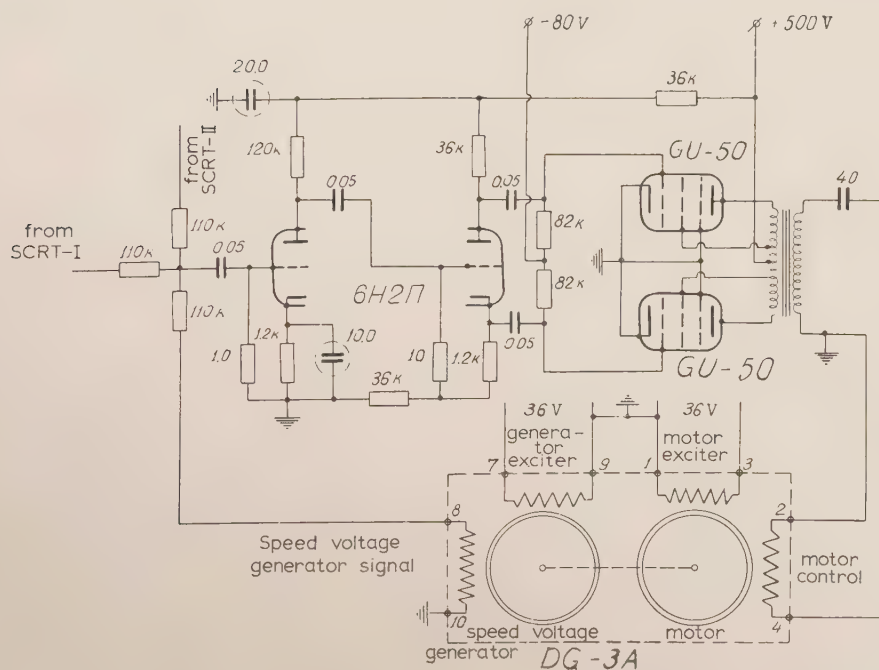


Fig. 6. Motor control circuit.

limits of 2–4% at any rotation angle of the photoelectric sensing due to careful adjustment of the photosensitive device to incident light. This provides an accuracy about 1–2 microns (on the film) of the coordinate measurements, the half width of the slit being 0.5 mm. Since the light signals are detected by four different phototubes, one may expect that their sensitivity changing differently with time, this will cause disturbances in the balance of the photosensitive device. The experimental work with the apparatus, however, has proved that during a day's work no disturbances of the balance occur. Practically, readjustment of the signals has been necessary once in 2–3 or even more days.

The measuring stage is driven by means of motor-generators "DG-3A" with frequency 400 c/sec. The use of speed voltage generators enables to introduce a negative rotation velocity feedback. The feedback coefficient has been chosen equal to 0.9. This has provided the necessary linearity of the control characteristic ($\approx 2\%$).

The motor control circuit is shown in fig. 6. To improve the shape of the signals to be applied to the motors, superlinear amplifier circuits²⁾ are used. The two step direct connection reducing gear has a much smaller momentum of inertia as compared to that of the motor. This is a necessary condition for a good rotational speed of the system. The gain of the parallel error-signal circuits has been selected so that on the sharpest tracks self-oscillations would be absent. Under these conditions the tracking unit has $\approx 20\%$ supercontrol. Under optimum conditions continuance of the transitory process in closed circuit has been $\approx 25 \mu\text{sec}$ long.

A switch on the control panel enables the operator to change the tracking velocity during the run, to make stops and change tracks. The rotation angle of the photosensitive device can be changed by the operator during the run as well as during the stops. It is possible to turn out the angular incongruity signals or all the error signals completely. The tracking unit can switch from dark tracks on bright background to bright tracks on dark background, if necessary.

²⁾ See, e.g. V. Labutin, "Radio" N II (1958) 42.

³⁾ J. Guild, *The Interference Systems of Crossed Diffraction Gratings* (Clarendon Press, Oxford, 1956).

4. The Numerical Reference System

The linear displacements are measured by means of diffraction gratings with a 10 micron pitch. The effect of Moiré fringes is used, arising when two diffraction gratings turned by small angle to each other are superimposed³⁾. If such a pair of gratings is illuminated with a parallel light beam, a set of dark and bright fringes appears in the space behind. The distance between two neighbouring fringes of equal brightness (the pitch of Moiré fringes) depends on the angle by which the gratings are turned to each other, and it can reach a remarkable value. If the gratings move relative to each other, the Moiré fringes are displaced so that to the displacement of gratings by one pitch (10 microns in our case) corresponds a displacement of the Moiré fringes by one pitch (i.e. several mm). To convert the continuous linear displacements into successive pulses the alternation of brightness of the fringes is registered by four photocells arranged at a distance equal to $\frac{1}{4}$ of the Moiré fringes pitch from each other. Such a disposition of the photodiodes forms a convenient and reliable detection unit of the direction in which the fringes are moving; this also provides an accuracy of grating relative displacement measurements equal to $\frac{1}{4}$ of grating pitch, i.e. 2.5 microns in this case.

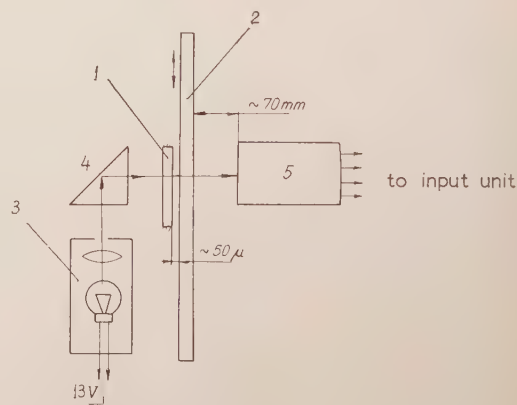


Fig. 7. Optical scheme of the reference system: 1. index grating; 2. scale grating; 3. illuminator; 4. deflecting prism; 5. array consisting of 4 photodiodes.

The layout of the measuring device is shown in fig. 7. The index grating is rigidly mounted together with photodiodes and illuminator. The other one, scale grating, is connected with the

age, displacement of which is being measured. The scale is 150 mm long. The index grating is made on a glass disc 40 mm in diameter, which is placed into a rotating frame. Separation of the two gratings is adjusted to be equal to 50 ± 2 microns.

differential amplifiers. Whenever the illumination of the corresponding pair of photodiodes becomes equal, the trigger state is changed. Pulses (negative as well as the positive ones) generated by the triggers as they switch from one state to another, are

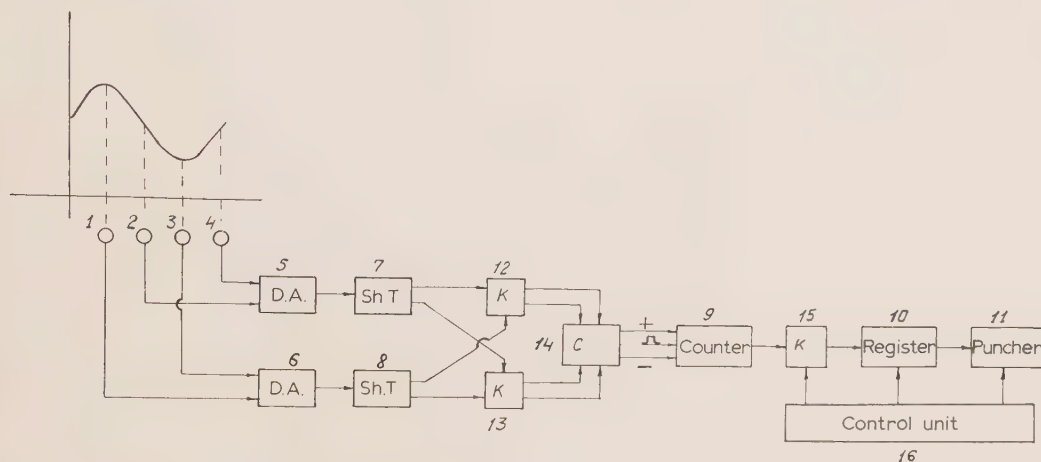


Fig. 8. Block diagram of electronic circuit of the reference system:

1, 2, 3, 4. photodiodes; D. A. differential amplifier; Sh. T. Schmidt trigger; K. valve system; C. signal collectors.

As photoelectric sensing photodiodes of the type "FD-2" are used, assembled in a packet. The height of the packet (between the centers of the outer diodes) is about 6 mm; according to this the pitch of Moiré fringes has been arranged equal to 8 mm. The photodiodes are selected to have approximately identical sensitivity and minimum dark current as well as possibly equal temperature dependence of the dark current.

An incandescent lamp "SM-25" (13 V, 25 W) is used for illumination. Diameter of the illuminator lens $d = 40$ mm, focal length $f = 60$ mm.

Due to careful adjustment of the gratings, illuminator and cylindric converging lens which is placed between the gratings and photodiodes, it has been possible to achieve an output signal from the photodiodes about 3-5 V at a level of about 12 V. The block diagram of the reference system is shown in fig. 8. Signals from the photodiodes 1-4 are coupled and fed into differential amplifiers 5 and 6, the output signals of the amplifiers corresponding to the illumination difference between photodiodes 1, 3 and 2, 4, respectively. Two Schmidt triggers 7 and 8 are controlled by the

used as reference pulses and delivered to a reverse counter 9. The hysteresis of the Schmidt triggers protects the system from false counting in the case that the system gets shaken. The state of the other trigger (its potentials) at the same time by means of a valve system (fig. 9) determines whether the pulse coming to the counter is going to be added or subtracted.

The circuit of the reverse counter is shown in fig. 10. Various trigger cells are interconnected by a valve system. Depending on whether the pulses should be added or subtracted one or another valve group opens (K0, K2, ... K30 or K1, K3, ... K31).

In order to punch the coordinates on perforated tape during the run without interposing the counters, every new number recorded on the reverse counters immediately is registered in a memory cell 10 (fig. 8). The memory circuit is represented in fig. 11). Every trigger cell of the memory circuit is connected with the corresponding counter cell by a valve set K1, K2, ... K16. Washing of the old number and recording of a new one occurs while the valves are open. While the coordinates are being

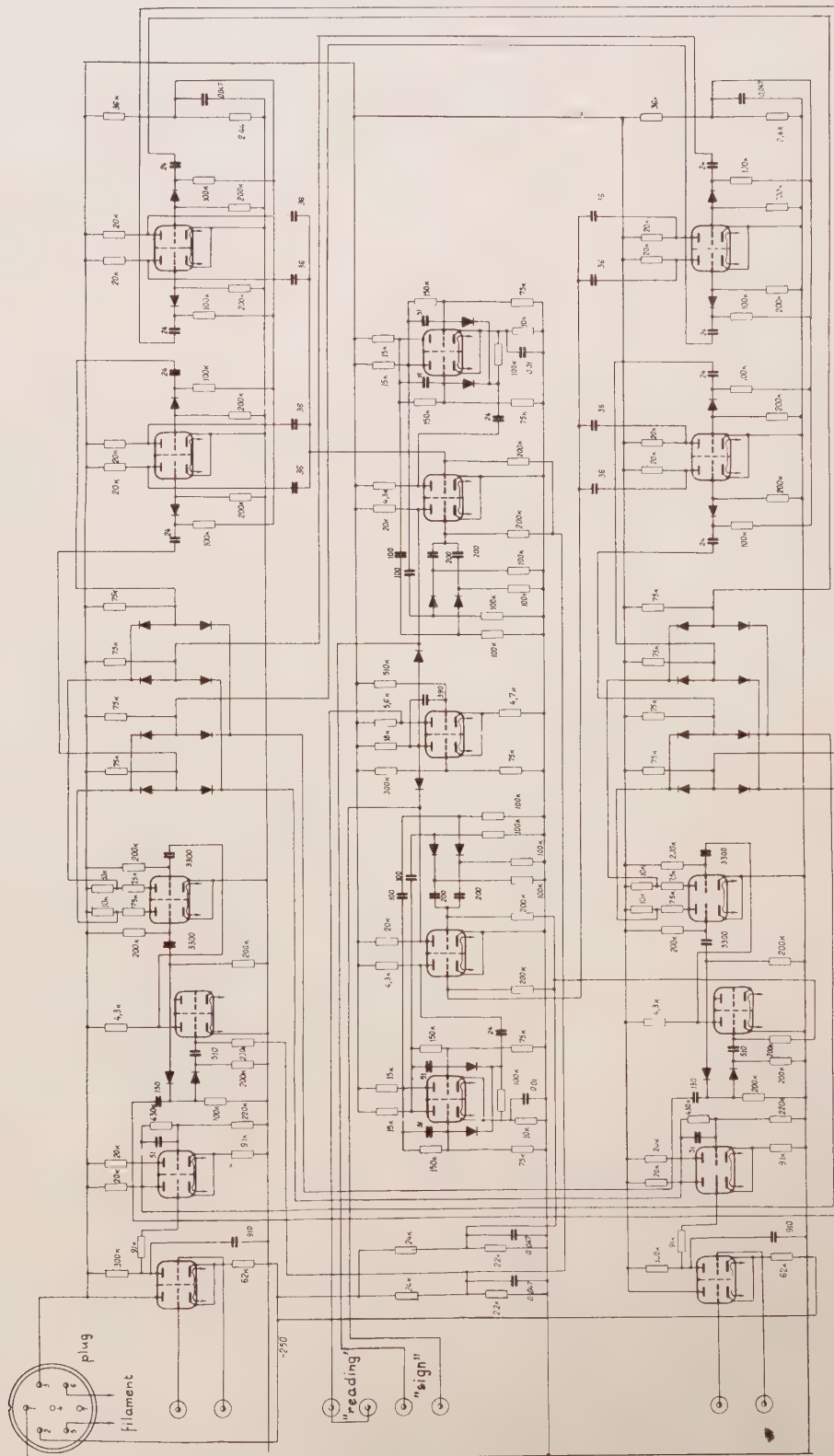
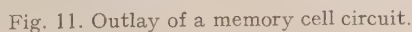


Fig. 9. Circuit of the input of the reference unit.

Besides the one-kick coordinate punches commenced by the operator each time, a special device provides automatic punching of coordinates after a certain interval of path along the track is passed, the path length being settled by the operator previously. Punching occurs only if the error-signals are absent and if the photosensitive device is not looking to a gap in the track.



In addition to the track coordinates, other information as the zone index and number, track number, frame number etc. has to be recorded on the tape. To make this work easier and to decrease the probability of errors a special device is provided. All the information numbers are selected by the operator in advance by means of switches which commutate different contacts of a step-type searcher. A push-button starts the searcher simultaneously with the perforator, so that the marks would be punched on the tape at the necessary places as the searcher moves on. The "SI 25/8" type searcher is used in the apparatus.

5. Conclusion

Employment of the apparatus has proven that it permits to increase the speed of photograph evaluation by several times as compared with manual examination under a microscope. Thus, to measure

20 coordinates on each track of a 4-beam star (two frames) by means of the automatic apparatus requires 10–15 min. On a measuring microscope it would require 4–6 hours. The mean-square error of measurements on the automatic apparatus is 3–4 microns. It has to be noticed that to employ the advantages of automatic photograph evaluation on "APT" completely, one has to use fast-acting computers producing several thousand operations per second. To compute space coordinates and curvatures of the tracks e.g. by "Ural-1" requires 5–8 times as much time as measurements of the coordinates on the automatic apparatus.

Acknowledgement

The authors would like to express their gratitude to Prof. V. P. Dzhelepov for his constant interest and aid to this work.

APPLICATION OF SURFACE BARRIER SOLID STATE IONIZATION CHAMBERS TO MEASUREMENTS ON FISSION[†]

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Gold-silicon surface barrier detectors $\sim \frac{3}{4}$ " in diameter have been made and used in several experiments on the fission process: (1) A pair of detectors, with detecting surfaces parallel and facing each other at a distance comparable with their diameters and with U^{235} deposited on one detector, has been used to measure the mass distribution in fission with thermal neutrons for several values of the total kinetic energy of the fragments. Values as high as 500 to one for the ratio of asymmetric to symmetric fission were obtained. (2) A similar arrangement,

but including a third detector, was used for the measurement of the mass distribution in ternary fission. (3) The ratio of ternary to binary fission for U^{235} fission with thermal neutrons was determined. (4) The variation of the fission cross section of U^{233} with energy was measured for neutron energies in the range 6 eV to 19 eV. The usefulness of solid state detectors and their superiority over most other types of known detectors for these measurements is clearly evident.

1. Introduction

The success of G. A. Dearnaley and A. B. Whitehead¹⁾ at Harwell with surface barrier gold-germanium and goldsilicon detectors indicated that these would be very useful devices for measurements on the fission process. In order to explore this application, measurements were carried out on the mass distribution in both binary and ternary fission, on the ratio of ternary to binary fission, and on the fission cross section in the resonance region.

The distinguishing features for the present work are (a) use of detectors of much larger areas ($\sim \frac{3}{4}$ " in diameter) to achieve the higher counting rates usually required in work with neutrons, and (b) mounting of the detectors in such a way as to minimize the amount of scattering material present, a factor which is particularly important in the measurement of the energy dependence of the fission cross section in the resonance region.

2. Fabrication of the Detectors (fig. 1)

Single crystals of silicon, $\sim \frac{3}{4}$ " in diameter and of thickness ranging from 0.015" to 0.040" and of ~ 50

ohm-cm resistivity, were obtained from Semiconductors Ltd., Swindon, Berks., England. After an etching in a CP4²⁾ type solution for ten minutes,

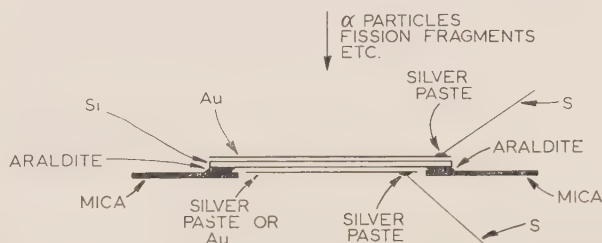


Fig. 1. Details of detector units. S = silver-gilt galvanometer suspension strip $0.001" \times 0.010"$. The gold (Au) layers of $\sim 100 \mu\text{g}/\text{cm}^2$ thickness were produced by evaporation. All of the data of this report were taken with detectors having a layer of silver paste, with the lead embedded in this, on the bottom (back) side.

washing in distilled water, and drying, the crystals were mounted with epoxy-resin on thin sheets of mica (0.003" to 0.005") over holes in the mica of the same shape as the crystals, but $\sim \frac{1}{8}$ " smaller, and baked at 100° for $\sim$ one hour to set the araldite. After a second, one-minute etch, gold was evaporated onto the larger exposed surface. Electrical

¹⁾ G. A. Dearnaley and A. B. Whitehead, unpublished Harwell reports.

²⁾ Conc HNO_3 , two parts; glacial acetic acid, one part; 40% HF, one part. Apparently, other proportions may be used with success.

[†] This work partially supported by the U.S. Atomic Energy Commission.

^{††} Work performed while the author was at Harwell on leave from Columbia University. This is also his present address.

connections were made to the front goldcoated surface and the back gold-free surface with silver gilt galvanometer suspension strips attached with silver paste.

The resolution of these detectors was about 20% or ≈ 1 MeV for the 4.76 MeV α particles from U^{234} . This is adequate for most work on fission fragments since the resolution from source thickness and other effects is usually worse.

After the investigations of this report had been completed, detectors giving about 5% resolution for the 4.76 MeV α 's were made by evaporating gold on to the back surface also before attaching the lead with silver paste.

The apparently poor resolution of these detectors is primarily due to their large size, since, under the conditions of operation, the preamplifier noise is independent of detector capacitance, but the signal is inversely proportional to the capacitance, which is proportional to the area. Thus the noise to signal ratio is directly proportional to detector area.

3. Mass Distribution in Binary Fission

In order to determine the mass distribution in fission, it is necessary to make simultaneous measurements on both fragments in the fission event. For this purpose, the two detector arrangement shown in fig. 2 was constructed, mounted in an evacuated chamber, and exposed to the thermal flux of $\sim 10^6$ neutrons/cm²/sec from hole E2/10 of the reactor BEPO. The U^{235} was evaporated directly on to one of the detectors, eliminating the supporting thin film which is necessary for corresponding arrangements using ionization chambers, gas scintillators, etc.³⁾ This arrangement is then, the equivalent of the back-to-back fission chamber used, for example, by Brunton and Hanna⁴⁾. The collimation usually required to select only those fragments emerging at reasonably large angles from the plane of the film of uranium is achieved here simply by the separation of the two detectors.

³⁾ The suspicion arises frequently that the U^{235} diffuses into the silicon so that the U^{235} layer becomes effectively thicker than desired. For this reason, it may become necessary again to deposit the U^{235} on thin films. The remaining advantages of this arrangement, however, remain.

⁴⁾ D. C. Brunton and G. C. Hanna, Can. J. Research **28A** (1950) 190.

In addition, no area, and hence counting rate, is lost in the opaque spaces required for the collimation, and the degree of collimation is readily adjustable by varying the separation of the detectors.

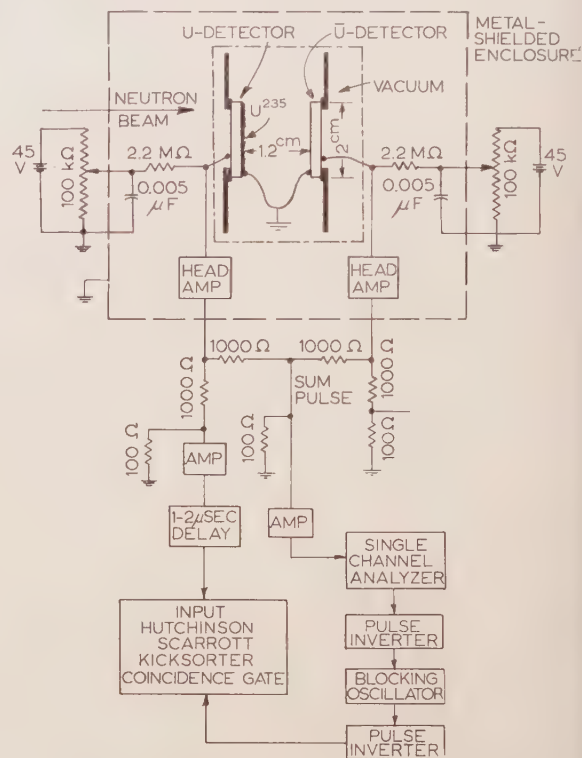


Fig. 2. Schematic diagram of the set-up for measuring the mass distributions. U and $\bar{U}$ refer, respectively, to the detectors with and without uranium coating. The counting rates with no coincidences were, for the pair of detectors used in the mass distribution measurements,

$\bar{U}$ = Si No. 6	18 α counts/sec
	14.5 fission counts/sec
U = Si No. 8	77 α counts/sec
	70 fission counts/sec

for 45 $\mu\text{g}/\text{cm}^2$ of U^{235} over a 1.5 cm diameter area on Si No. 8 and for neutrons from hole E 2/10 of the reactor BEPO with an estimated flux of 10^6 neutrons/cm²/sec.

The responses of the two detectors to the same fission fragment spectrum were matched by observing the kinetic energy distribution through the adding circuit with each detector disconnected in turn and adjusting the bias voltages on the detectors. The fission fragment energy spectra thus observed in detectors Si No. 6 and Si No. 8 are shown in fig. 3. The spacing collimation obtained for S

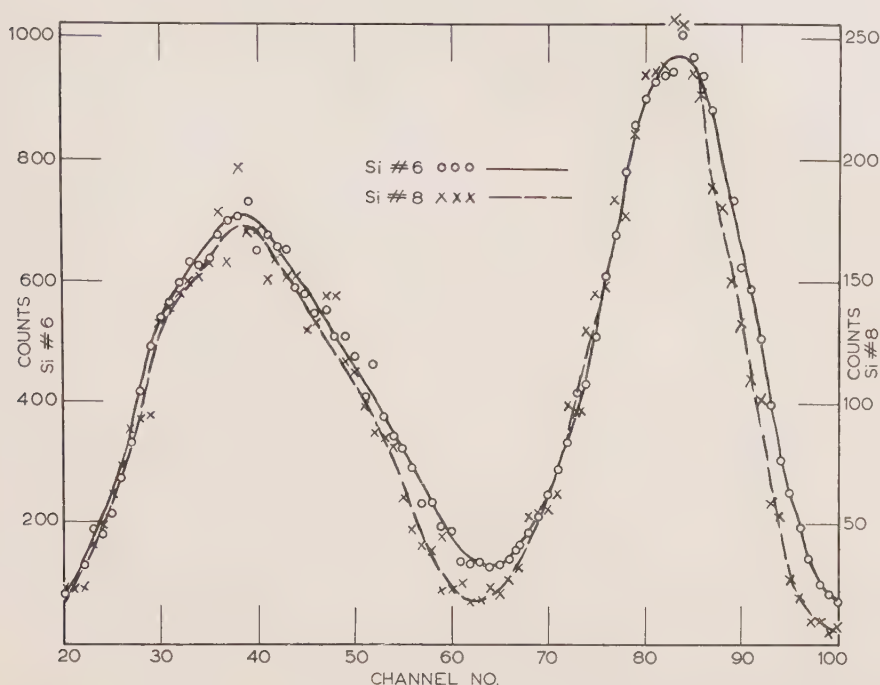


Fig. 3. Fission fragment energy spectra taken through the adding circuit of fig. 2, with each detector disconnected in turn and used to gate the kicksorter. In this way, only fission fragments entering detector Si No. 8 (XXX) approximately normal to the surface were accepted, hence avoiding those fragments which lose excessive energy going through the uranium and gold at a shallow angle. The data on Si No. 6 (ooo) were taken in the same way for symmetry, although the coincidence technique was not necessary. Detector Si No. 8 should have had a little more voltage on it to bring its fission spectrum into better alignment with that of Si No. 6. This particular re-

alignment was made after the mass distribution data presented in this paper were taken. Subsequent results on the mass distribution showed considerable difference in the number of counts under the light and heavy peaks, an effect probably caused by the slight misalignment. The earlier results of this paper were apparently done with better alignment, although the energy spectrum data were not recorded. The ratio of high energy peak to valley is $7\frac{1}{2}$ to 1 for Si No. 6 and 13 to 1 for Si No. 8. Dr. Terry Gooding⁷⁾ states that in an identical system he obtained a value of 17/1 for the ratio of high energy peak to valley, but that this deteriorated to values comparable with the above after a few months.

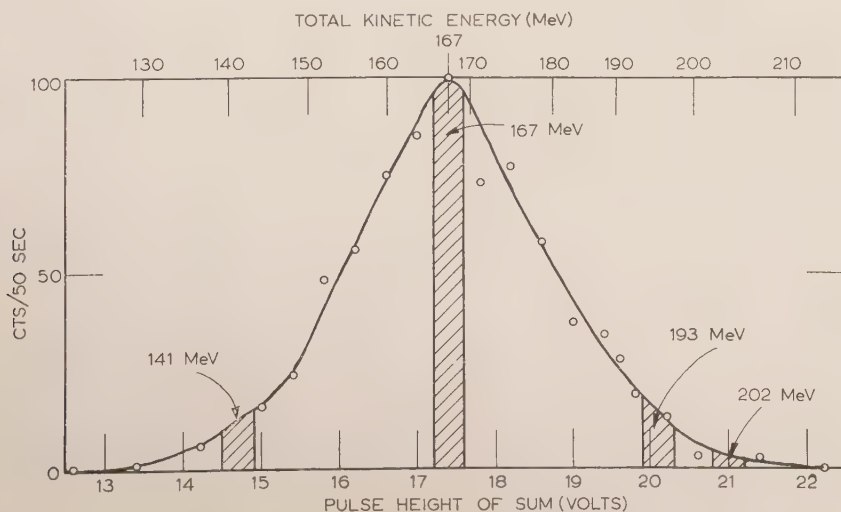


Fig. 4. Count rate as a function of pulse height in the single channel analyzer (of fig. 2) which accepts the sum of the pulses from the two detectors. The calibration in MeV is based on the

assumption that the peak corresponds to 167 MeV. The full width at half maximum is 26 MeV. The shaded areas indicate the regions accepted in the mass distribution results shown in figs. 5-8.

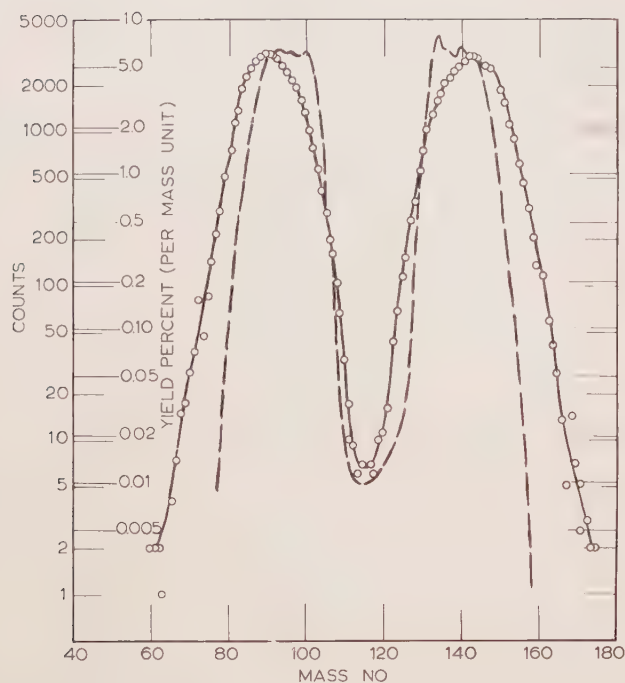


Fig. 5. Mass distribution for total kinetic energy at the peak of the E_K distribution (167 MeV). A peak to valley ratio of ~ 500 is obtained. The radiochemically determined distribution, for all events, is included for comparison (---).

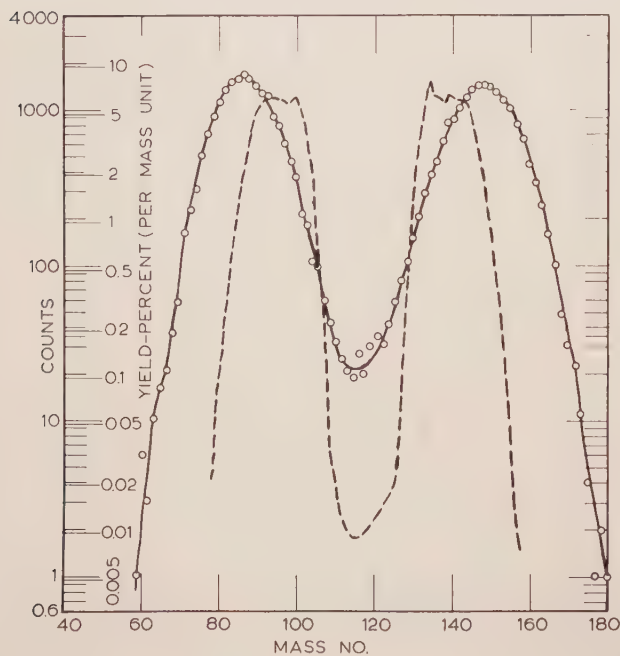


Fig. 6. Mass distribution near the low energy end (141 MeV) of the total kinetic energy distribution. A peak to valley ratio of ~ 70 is obtained. The radiochemically determined mass distribution, for all events, is included for comparison (---).

No. 6 is applied to Si No. 8 by accepting only those fragments which are in coincidence with the complementary fragment in Si No. 6. The peak-to-valley ratios observed have been used as a measure of the quality of the measurements. The result, 13/1, for detector Si No. 8 is better than reported in the literature. The results for Si No. 6 are not so good presumably because of edge effects in Si No. 6 and/or a thicker gold layer in Si No. 6 (Edge effects in Si No. 8 have been avoided by coating with uranium only to within $\sim \frac{1}{8}$ " of the edge.) The energies of the most probable high and low energy peaks (calibrated with respect to the U- α 's) are 91 MeV and 55 MeV, and their ratio = 1.65. The expected values, based on earlier work corrected for ionization defect when necessary, are 100 MeV, 67 MeV, and 1.50 respectively. These discrepancies may have been caused by loss of energy in the gold and uranium coatings, although an estimation of these effects does not appear to account entirely for the discrepancy. Diffusion of the U^{235} into the silicon³) may be a significant factor here.

Considering only the conservation of momentum in the act of fission and ignoring as small the effect arising from neutron and γ -ray emission, it can readily be shown that

$$\frac{M_1}{M_1 + M_2} = \frac{E_2}{E_1 + E_2} = \frac{E_K - E_1}{E_K}$$

where M_1 and M_2 are the masses of the fragments, E_1 and E_2 their respective energies, and $E_K = E_1 + E_2$. Since $(M_1 + M_2) \approx 233.5$ ($235 + 1 - 2.5$), the mass distribution can be obtained from the energies of the fragment by adding the energies of the two fragments and performing the indicated division electronically⁵). An electronic set-up to do this was not constructed. Instead, the electronic arrangement of fig. 2 was used, where the sum energy E_K was gated through a single channel analyzer with $\Delta E_K = 4$ MeV for various values of E_K , and an energy distribution obtained for those events in coincidence. The result is proportional to the mass distribution for a small range of the total kinetic energy.

⁵) L. W. Roeland, L. M. Bollinger and G. E. Thomas, Proceedings of the Second United Nations International Conference on the Peaceful Uses of Atomic Energy, P/551, 15 (1958) 440.

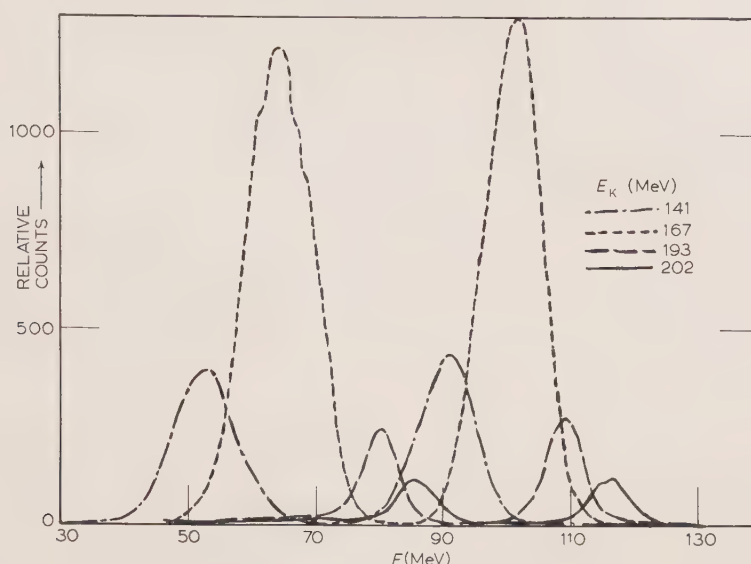


Fig. 7. Composite plot of data taken at four different values of the total kinetic energy (E_K) normalized to the same counting period as for $E_K = 167$.

Fig. 4 shows the distribution of E_K and the 4 bands selected for measurement.

Fig. 5 shows the mass distribution obtained at the peak of the E_K distribution. A peak-to-valley (asymmetric to symmetric fission) ratio of ~ 500 is obtained. The corresponding mass distribution obtained by radiochemical means, which includes all events for all E_K , indicates a peak-to-valley ratio of 650⁶). This difference may be due to the difference in the range of E_K accepted in the two types of measurements, but is more likely to be caused by the energy loss by the fragments in the various coatings and by the lack of perfect matching of the responses of the two detectors. It should be noted that, in all of these mass distribution curves, the count rate goes essentially to zero on the high and low sides of the distributions, so that there is no possibility of an over-all constant background.

Fig. 6 shows the results for the smallest value of E_K investigated (141 MeV) giving a peak-to-valley ratio of ~ 70 . Thus the relative amount of symmetric fission appears to increase at lower total kinetic energy.

Details of the results on the high energy side of the total kinetic energy distribution are not included here because of irregularities in the results. However, further investigations by Dr. Terry Gooding⁷) on the same set-up indicate a decrease in

the relative amount of symmetric fission at high total kinetic energy.

Figs. 7 and 8 show composites of the data, taken at the four values of E_K shown in fig. 4, with

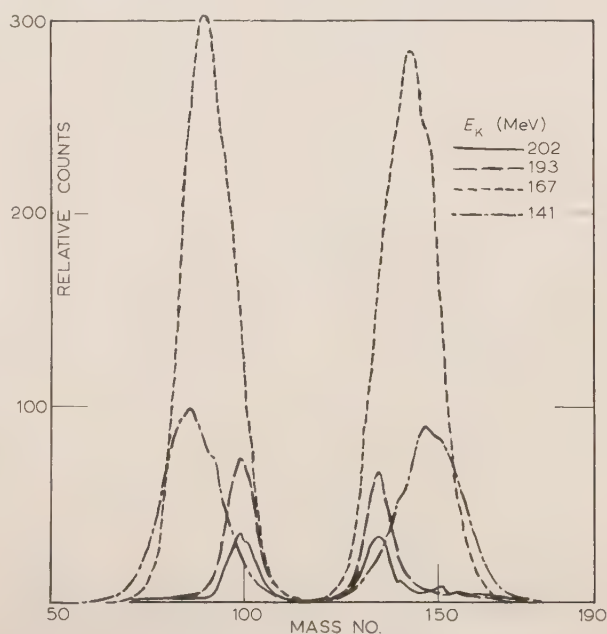


Fig. 8. Same as for fig. 7, except that the energy abscissa has been converted to a mass scale.

⁶) S. Katcoff, Handbook of Nuclear Engineering (Addison-Wesley, 1957).

⁷) Dr. Terry Gooding, private communication.

energy and mass abscissas respectively. While earlier results (e.g. those of Brunton and Hanna⁴) gave information corresponding to that shown in figs. 7 and 8, none of them were of sufficient statistical accuracy to give information on the relative amount of asymmetric-to-symmetric fission as is the case here.

4. Mass Distribution in Ternary Fission

D. L. Hill⁸) has suggested that the mass distribution in the most probable form of ternary fission, i.e. with α emission, may be essentially symmetric, unlike the case of binary fission. In order to perform such a measurement, the three detector arrangement of fig. 9 was set up. If detectors $\bar{U}$ and U only are considered, the arrangement can be used to

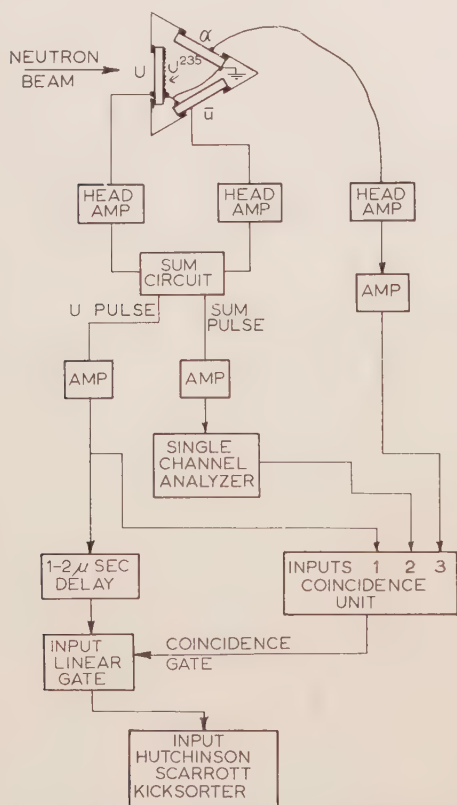


Fig. 9. Set-up (schematic) used in the determination of the mass distribution in ternary fission.

measure the mass distributions exactly as with the set-up of fig. 2, except with somewhat poorer geometry. If, now, a coincidence is required with detector " α ", the desired mass distributions in the

case of accompanying α emission would be determined provided there were no accidental coincidences. According to the count rates observed, the rate of accidental coincidences should have been negligible compared with the number of coincidences actually observed. Fig. 10 shows these results. Because of the low count rate and the limited time available for the measurement, the statistics are very poor, but the indications are that the mass distribution is similar to that in the case of ordinary fission.

5. Ratio of Binary to Ternary (α) Fission

The ratio of yields of binary to ternary fission has been measured to be in the range 250 to 500⁹), but there appeared to be disagreement as to the exact value. It was thought that the arrangement of fig. 2 would be suitable for this type of measurement. Under ideal conditions, with the detector "looking at" the uranium from some distance from it, there should be a count-free interval between the ordinary α particles and the lowest energy fission fragments in binary fission. α 's from ternary fission should show up, then, in this region. Assuming complete lack of correlation in direction with the incoming beam, the geometry for α 's and fission fragments would be identical, so that counts of the number of ternary α 's and fission fragments should yield the ratio directly. Numerous attempts to observe a definite group of α 's (with a maximum energy 26 MeV) and a count free gap to the lowest energy fission fragments (~ 40 MeV) always resulted in a continuum of events larger in number than the expected number of α 's from ternary fission.

In an alternative attempt to measure the same ratio, an 8.2 mg/cm² aluminum foil was interposed between the detector and the uranium. Fig. 11 shows the results with and without cadmium in the neutron beam. An extra peaked distribution is seen, attributed to the α 's sought. Combination with the observed number of fission events normalized to the same period of time gives a value of 330 ± 40 for

⁸) D. L. Hill, Proceedings of the Second United Nations International Conference on the Peaceful Uses of Atomic Energy, P/660, 15 (1958) 244.

⁹) W. J. Whitehouse, Progress in Nuclear Physics, Vol. 2 (Pergamon Press, Ltd., 1952) p. 120.

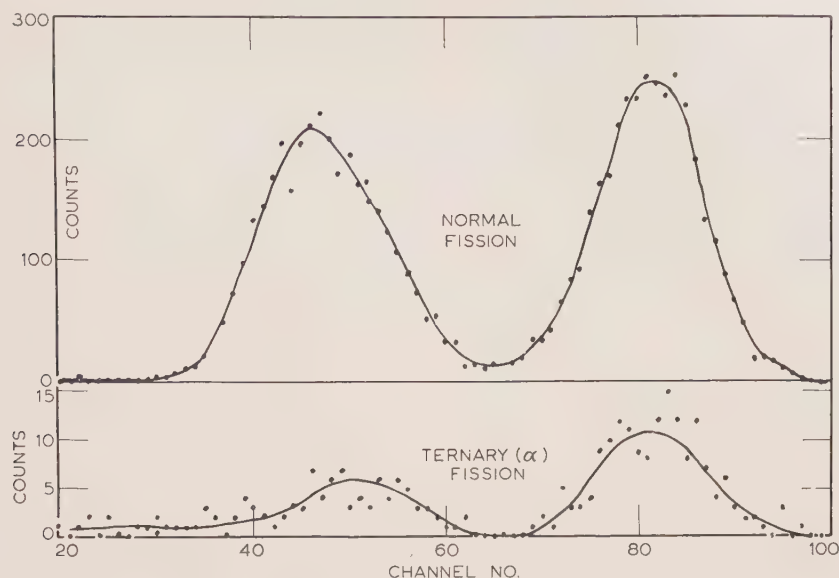


Fig. 10. Mass distribution in ternary fission compared with the distribution in normal fission for a total kinetic energy of 167 MeV. Both were taken with the arrangement shown in fig. 9. There appears to be no pronounced difference.

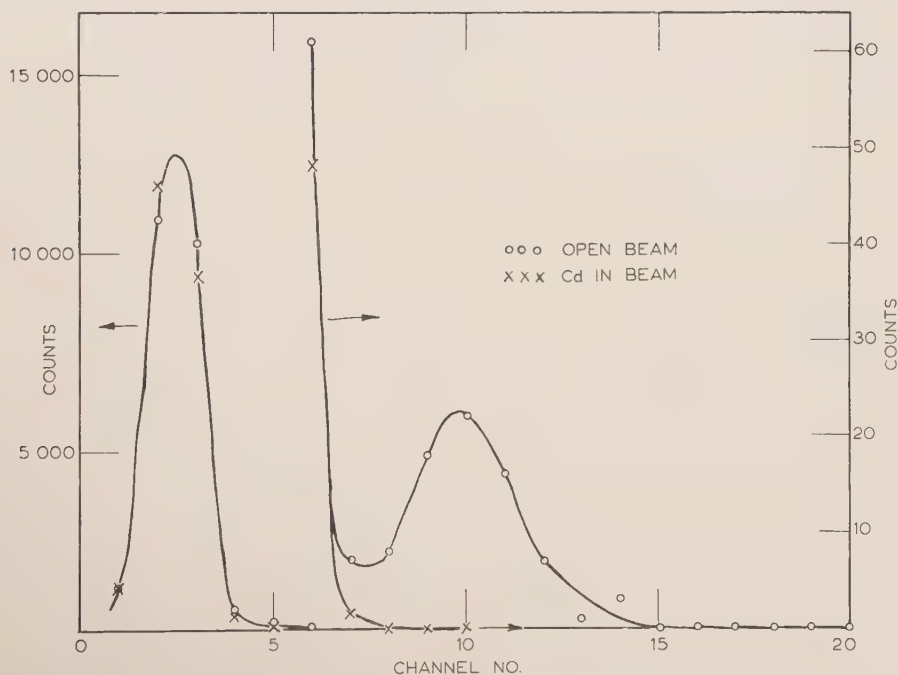


Fig. 11. Determination of the rate of α emission in ternary fission. The arrangement of fig. 2 was used with a Au-Si detector as $\bar{U}$ except that (1) the U detector was used only as a source backing, and (2) an 8.2 mg/cm² aluminum foil was placed between source and detector. This amount of absorption stops all fission fragments and all α particles less than ~ 6.6 MeV. Thus, reaching the detector are the 6.774 MeV α 's from Po^{216} , producing an unobservedly small effect, the 8.776 MeV α 's from Po^{212} , presumably producing the large peak seen in both open beam and cd-in-beam runs, and the α 's from ternary fission

causing the peak not observed in the cd-in-beam run. (The Po α 's appear to arise from a small contamination of U^{232} and its daughter products in the U^{235} deposit.) The open beam run (ooo) lasted 26 hours, while the cd-in-beam run (XXX) lasted for 15½ hours, and was normalized to 26 hours before plotting) 79 events are attributed to α 's from ternary fission. The observed fission rate without the aluminum absorber was 568 counts/1000 sec. Taking into account the fact that fission events are detected with twice the efficiency as are the α particles, the ratio of normal fission to that with α emission is 330 ± 40 .

the ratio of fission events without α emission to those with α emission.

Since both types of measurements on ternary fission were not repeated and subjected to thorough tests, the results should be accepted only tentatively. They were performed to explore the measurements possible with these detectors.

Also, their high speed makes possible the use of shorter resolving times, reducing the amount of α particle pile-up in the case of isotopes like U^{233} and Pu^{239} , which have very high α activities.

To investigate this application, a measurement was made on U^{233} at a 15 meter flight path of the 28 MeV linear electron accelerator. Only $2\frac{1}{2}$ hours

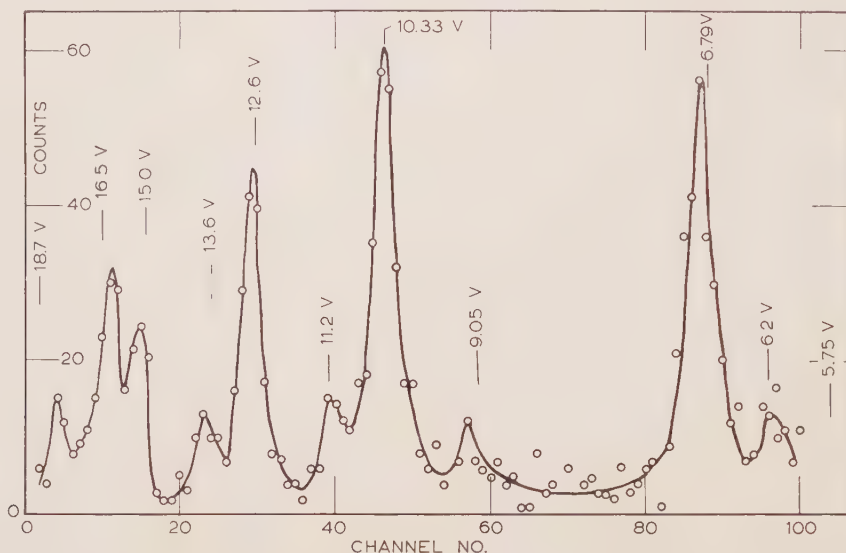


Fig. 12. Counts from a 1.3 mg U^{233} sample, $\sim \frac{1}{4}$ " from Au-Si detector No. 6, 15.46 m from the booster of the 28 MeV electron linear accelerator, taken with 2 μ sec gates and 260 μ sec delay.

Energy values from BNL-325, 2nd edition, are listed, based on a calibration using the levels at 10.33 eV and 6.79 eV to normalize the flight distance and delay time.

6. Measurement of Fission Cross Section as a Function of Neutron Energy

The measurements described above make use of the property of these detectors that pulse height is proportional to the energy of the fission fragment to study details of the fission process. The use of these detectors is also advantageous in the more straightforward process of detecting the occurrence of fission for the purpose of measuring fission cross section as a function of neutron energy. The possibility of making detector arrangements with a minimum of scattering material present is of particular advantage here. Their compactness makes possible the inclusion of much more fissionable material in the beam, so that count rates, almost always a problem here, may be increased.

¹⁰⁾ R. G. Fluharty, M. S. Moore and J. E. Evans, Proceedings of the Second United Nations International Conference on the Peaceful Uses of Atomic Energy, P/645 15 (1958) 111.

of data-taking gave the results of fig. 12, showing at most 60 counts in the peaks of the resonances. In spite of this, the results are extremely clean with no significant bumps on the high energy side of the levels arising from the effects of scattered neutrons. There appears to be little background, since the regions between levels correspond to those of the MTR results¹⁰⁾. All of the levels, except the one at 9.05 eV are clearly seen in these results.

It would appear that the use of solid state detectors is superior, in general, to other currently known detectors, in the measurement of the energy dependence of the fission cross section.

7. Comments

The usefulness of solid state detectors in measurements on the fission process is clearly evident. In many cases they are superior to other types of detectors.

One improvement in construction may be noted here relating to the evaporation of the gold. The amount of gold deposited on the detectors used in the measurements of this report was judged "by eye" resulting in wide variations in the amount of gold deposited. Subsequently, a glass plate was fitted with electrodes about 1 cm apart and placed near the detectors in the evaporation equipment. As the evaporation proceeded, the decreasing resistance across the glass plate was observed and the evaporation stopped at a resistance ($\sim 10-45$ ohm) corresponding to the minimum acceptable amount of gold.

Only solid state detectors of the surface barrier type were studied in these investigations since the diffused p-n junction type¹¹⁾ was not available. The latter may be equally useful in fission measurements, although those which have been made are generally of small area and hence lead to low count rates.

Acknowledgements

The author is grateful to Professor W. W. Havens, Jr. and Professor C. S. Wu of Columbia University and to the U.S. Atomic Energy Commission for making possible the time spent at Harwell, and to Dr. E. Bretscher and Dr. E. R. Rae for making the stay at Harwell so profitable and enjoyable. The author is indebted to G. A. Dearnaley for much advice and assistance in the preparation of detectors suitable for the work presented here, and to Dr. Terry Gooding for his collaboration in some of the later stages of this work and for communicating the results of his subsequent investigations prior to publication. It is a pleasure to acknowledge the cooperation of Drs. R. Bird, J. Fraser, D. Gayther and D. Colvin, and Messrs. F. Brooks and N. Nobes in many phases of the experiments. Thanks are also due to Messrs. Hutchinson, Furby and Wilkinson for the preparation of the U^{235} and U^{233} deposits.

¹¹⁾ Nucleonics 18, No. 5 (1960) 98.

PILE UP REJECTION CIRCUITS

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The paper discusses the main pile up effects and the methods used for their rejection. Two new circuits are described which generate gates that interact with the signals to be registered every time a pile up effect is detected. The first circuit has a resolving time of 350 nsec and a pile up threshold level of 4 V,

when operated from the output of an ordinary amplifier. The resolving time and threshold level of the second circuit are reduced to 30 nsec and 1 V respectively, when operated directly from the dynode output. The scintillator used was NaI(Tl).

1. Introduction

In experiments where the expected event occurrence rate is low, the background rate becomes critical. The increase of the counting period above a certain value in order to improve the statistics is not recommended, mainly due to the problems of long-term stability of the electronic equipment. When the source life time is relatively short, further complications are encountered. The preferred solution is to increase the source intensity. Random counts are minimized by incorporating "conditions" on energy and/or time which have to be fulfilled for a count to be registered. Usually, discriminators and single channel analyzers serve for the energy spectrum.

In the time domain, fast and slow coincidence circuits are employed, but the electronic units preceding the final slow stages must be capable of dealing with the increased rate. The pile up effect due to the random occurrence of the pulses becomes an important factor when the rate is increased.

In those spectrum experiments where energy conditions only are used, piled up pulses may fulfil the required "conditions", and false records will be registered. Fig. 1 illustrates that case with single pulses normally below the minimum threshold.

In those "life time" measurements where time to amplitude conversion is employed, the pile up effect is most serious. The pulse height developed in the conversion unit is a function of the time difference between pulses and of the limiter-shaped output

pulse amplitude in the coincidence unit. The limiter shaped output pulse amplitude will be reduced if it was developed by a piled up pulse, and depend on the exact instant of occurrence (fig. 2). In this case, the converter output will not be a linear function of the time difference.



Fig. 1. The pile up effect with single pulses normally below the minimum threshold.

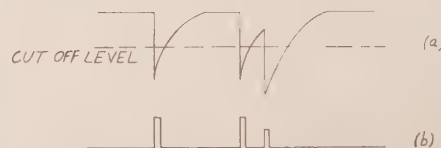


Fig. 2. The pile up effect at (a) the input to the limiter (b) the shaped limiter output.

The usual pile up elimination circuits are composed of RC or delay line clippers in the input circuits of the amplifiers, whose main purpose is to differentiate the pulses from their slow-varying decay. This system will function effectively when the decay time is much larger than the pulse rise time and for separation times, between pulses, longer than the clipping time constant.

This paper will discuss a different approach to pile up elimination circuitry. The resultant performance overcomes the above mentioned limitations. The circuits described will reject the ready-to-record signal whenever a pile up is detected.

2. Methods of Operation

The pile up rejection circuits generate gates which interact with the signals to be registered. As the interval of observation for pile up effects is usually few microseconds only, it is not practical to delay the input pulse to the energy units accordingly. The interaction will better be postponed to the later stages. The shape of the pulses developed by the discriminators and/or single channel analyzers and/or slow multi-coincidence units will considerably simplify the interaction circuitry.

Two modes of operation are possible: (1) A gate will be developed for every pulse when a pile up did not occur, (2) A gate is produced when a pile up did occur. In mode (1) the signal and "not piled up" gate are applied to a coincidence circuit. The rate of gate generation must be high and any pulse occurring during the dead time of the gate will not be registered. In mode (2) the signal and the "pile up" gate are applied to an anti-coincidence circuit. The gate rate will be considerably lower but the missing of a gate, due to dead time effect, will enable registration of a piled up pulse. However, the probability of successive piled up pulses to occur with such a time difference that a trigger will be developed during the short dead time of the gate is very low.

3. Principles of Operation

The pile up detector block diagram is shown in fig. 3. The differentiated input pulses, after amplification, are applied to a coincidence circuit and to a Sensitive Univibrator (SUV). The delayed output of the SUV is also applied to the coincidence circuit.

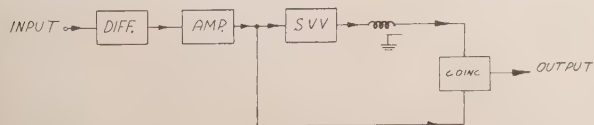


Fig. 3. The block diagram of the pile up detector.

The delay time of the line has to be longer than the differentiated input pulse width in order to avoid selfcoincidences. A coincidence output will be developed if a second pulse occurs during the active period of the SUV (observation period). The minimum resolving time is determined by the input

pulse rise time and the order of differentiation. The duration of the observation period is dictated by the time required for the maximum available pulse to decay below the minimum threshold regarded as a pile-up contributor. Of course not all pulses are of the maximum amplitude, hence the observation time is normally too long. The elimination of the unexploited time will increase the pile up detection rate. A possible improvement will be to use the actual input pulse itself as the delayed input to the coincidence circuit (fig. 4).

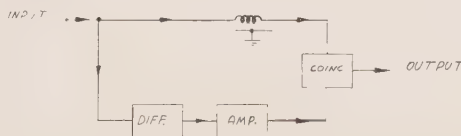


Fig. 4. The block diagram of the improved pile up detector.

When scintillators of relatively long decay time (NaI-Tl) are used, the exponential decay of the current pulse is only correct statistically. The instantaneous current amplitude fluctuations may appear as pile up pulses in case a high order differentiator is used for resolving time minimization. This effect sets, therefore, the practical minimum threshold level.

When short decay time scintillators (plastic), are used and the energy output is taken from a dynode with a "stretched" time constant, the raw short pulses may be applied either directly (without a differentiator) to the input of the circuit as shown in fig. 3 or in the manner shown in fig. 5.

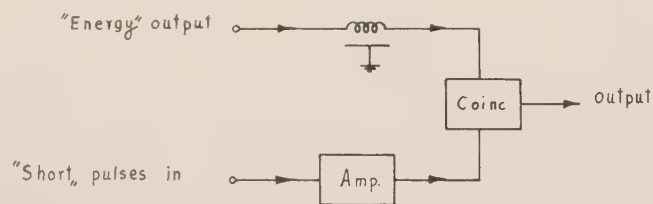


Fig. 5. The block diagram of the pile up detector when a short decay time scintillator is used.

The block diagram of the pile up rejection unit operating in mode (1) is shown in fig. 6. The SUV output is differentiated and its trailing edge pulse is applied to an anti-coincidence circuit.

The coincidence circuits trigger a Univibrator (UV) whose output is also applied to the anti-

coincidence circuit. In order to ensure that the anti-coincidence circuit will function properly whenever a pile up occurs during the observation period, the UV output pulse width has to be at least equal to the interval between the leading edge of the SUV output and the tail of its differentiated trailing edge. The Gate Univibrator (GUV) can be triggered by the anti-coincidence circuit only if the coincidence circuit did not function.

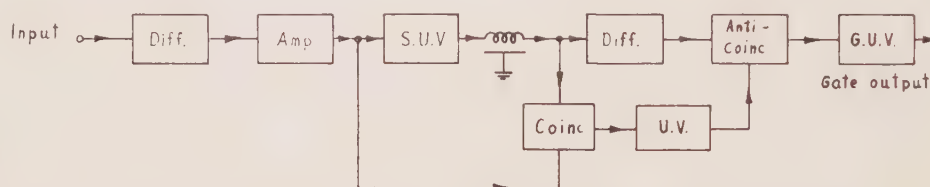


Fig. 6. The block diagram of the pile rejection unit operating in mode (1).

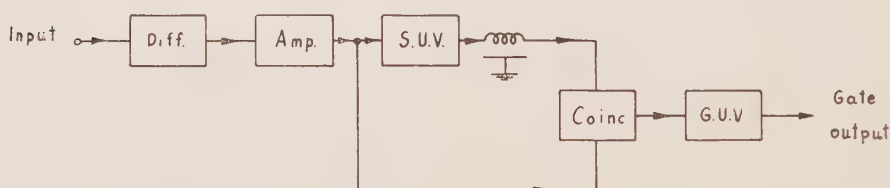


Fig. 7. The block diagram of the pile up rejection unit operating in mode (2).

The block diagram of the pile up rejection unit operating in mode (2) is shown in fig. 7. The GUV is triggered every time the coincidence circuit functions. It is clearly seen from fig. 7 that the complexity of the circuitry required is considerably reduced.

4. Practical Circuits

Two pile up rejection circuits operating in mode (2), ("Pile Up" Gate) with slow scintillators will be described. The circuit diagram of a unit according to the block diagram in fig. 7, is shown in fig. 8. The input differentiator is composed of C_1, R_1 , and the shorted transmission line TL_1 . The valve V_1 (E180F) serves as an amplifier and V_2 (E180F) as a limiter in order to compress the widely different input pulses. The SUV, V_3 (E88CC), is a cathode-coupled univibrator with a hold diode CR_1 . The coincidence circuit is composed of the diodes CR_2, CR_3 and CR_4 connected according to the Garwin configuration. The diode CR_5 is used for "singles"

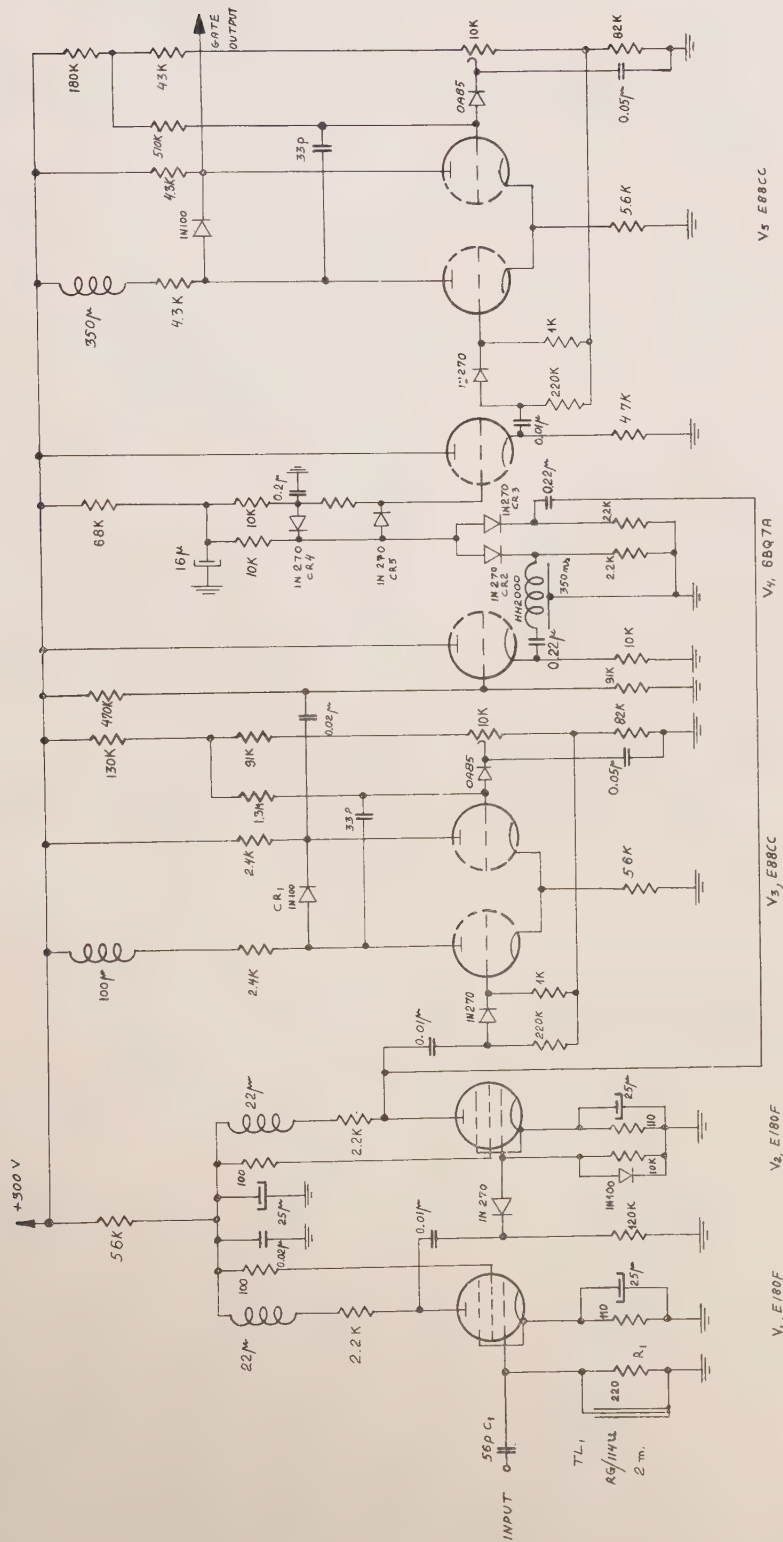
reduction. It is back-biased due to the forward voltage drop across CR_4 . The GUV, V_4 (E88CC) circuitry is similar to that of the SUV. If few pile-up rejection channels are used, the individual coincidence outputs may be coupled through isolating diodes to the same grid of the GUV (V_4).

Only one differentiator is used, hence the differentiated pulse width will equal the rise time of the input pulse which in our case is approximately

350 nsec. The SUV output must be accordingly delayed. The actual minimum resolving time will be shorter because the coincidence circuit will respond to the tail of the differentiated pulses. A "pile up" gate will be generated even if the leading edges of the differentiated pulses are closer than 350 nsec. The minimum resolving time will therefore depend on the relative amplitudes of the input pulses.

The negative slope of the input pulse following its peak develops, after differentiation, a negative pulse which reduces the instantaneous zero base line, that shift depending on the input pulse height. Its maximum value, for the highest permissible input pulse, must be lower than the positive differentiated pulse which corresponds to the maximum pile-up threshold. The pile up threshold level in this circuit is 4 V. A "pile up" gate will be generated even if the 4 V input pulse follows an 80 V pulse.

The observation period is 4 μ sec. The output of the GUV is a positive pulse of 30 V amplitude and 10 μ sec duration.



SYSTEME DE DETECTION DES PARTICULES ANALYSEES PAR UN AIMANT

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Reçu le 23 janvier 1961

A detection system is described which locates and counts particles analysed by a magnet. Ten proportional counters and a scintillation counter allow the detection of particles on twenty

independent bands of 5 mm width. A brief description is given of the associated electronics.

1. Introduction

L'utilisation d'un aimant pour l'analyse des particules diffusées présente l'avantage d'une bonne résolution en énergie. Par contre, les mesures sont plus longues que dans le cas de l'analyse par scintillations. On n'analyse en effet qu'une bande étroite du spectre énergétique à la fois. Pour diminuer la durée des mesures, il est important de disposer d'une détection qui analyse en une fois la plus grande portion possible de la focale image de l'analyseur. Dans notre cas, l'analyse se fait sur une bande de dix centimètres de long.

2. Disposition de la détection (fig. 1)

L'ensemble de la détection, fixé sur le bras de l'aimant analyseur et isolé électriquement de celui-ci, comprend trois parties. Ce sont, dans l'ordre de traversée par les particules: a. un compteur mesurant dE/dx ; b. compteur à dix sections dénommé décacompteur; c. un compteur mesurant l'énergie E .

2.1. COMPTEUR dE/dx (fig. 2)

Un scintillateur mince de CsI(Tl) (10 mg/cm^2 évaporé sous vide sur $200 \text{ } \mu\text{g/cm}^2$ de polystyrène) de $100 \times 20 \text{ mm}$ est placé perpendiculairement au faisceau. Un réflecteur en aluminium poli renvoie la lumière sur un guide placé devant la photocathode d'un photomultiplicateur EMI 6099 à grand diamètre (110 mm). Les particules alpha de 44 MeV perdant un MeV dans le scintillateur, après amplification nous obtenons des impulsions de l'ordre de 38 volts . La largeur à mi-hauteur est de 20% dont

une part est due à la dispersion liée au ralentissement. Un seuil réglé à 10 volts permet d'éliminer les protons et les deutons focalisés par l'aimant. Le système fournit une impulsion propre à effectuer des coïncidences avec les signaux délivrés par le décacompteur pour diminuer le bruit de fond.

2.2. DÉCACOMPTEUR (fig. 2)

Composé de dix compteurs proportionnels, le décacompteur est chargé de localiser les particules sur la focale image. Il a été construit suivant une technique étudiée par A. Papineau¹⁾ pour obtenir une grande stabilité de fonctionnement. Le décacompteur est constitué par une coque de laiton divisée en dix parties de 10 mm par des feuilles de nickel de $2/10$.

Chaque élément ainsi défini comporte un fil de molybdène. Pour assurer l'étanchéité de la coque et un bon isolement électrique du fil, les sorties se font par l'intermédiaire du passage métal-verre: Kovar-verre au molybdène. Le mélange gazeux utilisé est constitué par 5% de CO_2 et 95% d'argon, la pression totale étant équivalente à 40 mm de mercure. La coque est portée à une tension négative de 1500 V . Chaque fil peut être porté indépendamment à une tension variable de 0 à $+250 \text{ V}$ ce qui permet d'ajuster les seuils de comptage de chaque élément. L'efficacité des dix compteurs est ainsi rendue uniforme. La stabilité de l'ensemble s'est avérée suffisante pour que l'on n'ait pas eu besoin de corriger le réglage des seuils au cours des six mois de fonctionnement.

¹⁾ A. Papineau, J. Phys. Radium **12** (1951) 667.

2.3. COMPTEUR E (fig. 2)

Le compteur à scintillations E, placé derrière le décacompteur, mesure l'énergie restante des particules. Il est constitué par un monocristal de NaI(Tl) (dimensions: 110 mm sur 20 mm de haut et 2 mm d'épaisseur) suivi par un guide de lumière, entouré

par l'aimant, ou bien, pour améliorer la résolution de l'ensemble du système, on ramène la largeur de détection de 10 mm (décacompteur) à 5 mm en utilisant le compteur E comme codeur binaire. On opère alors de la manière suivante:

On place entre le décacompteur et le cristal E une

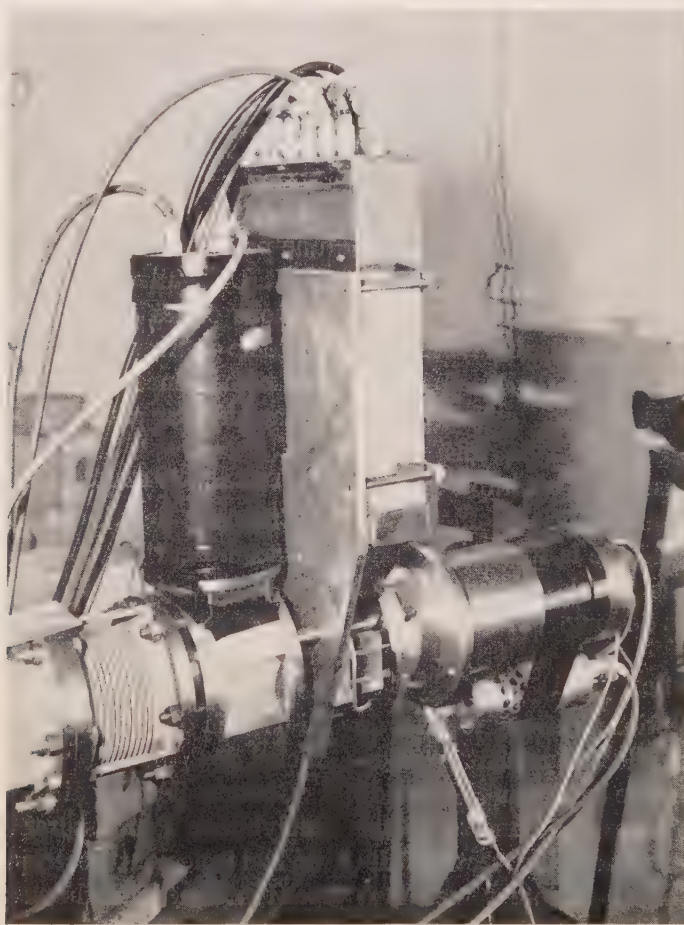


Fig. 1. Vue de l'ensemble de détection.

d'un réflecteur aluminé et placé contre un photomultiplicateur EMI no. 6099.

Après amplification on obtient une résolution de 6% pour des alpha de 44 MeV malgré la grande surface de détection. Le compteur E est utilisé suivant les besoins de deux manières différentes. Ou bien l'étude du spectre énergétique permet de déterminer sans ambiguïté la nature des particules détectées en combinant l'information d'énergie avec l'information de quantité de mouvement fournie

grille de tantale d'épaisseur 5/10 mm comportant dix ouvertures de 5 mm de large, 12 mm de hauteur, distantes de 5 mm chacune. La grille est calée de manière à occulter la moitié de chaque section du décacompteur. L'épaisseur du tantale étant suffisante pour arrêter les particules analysées, on détermine quelle est la portion de 5 mm qui est attaquée dans chacun des compteurs proportionnels suivant que l'on obtienne ou non un signal sur la voie énergie.

Traitement électronique des impulsions de détection

Le but essentiel de la détection consiste en un comptage des particules localisées. Aussi doit-on apporter quelques modifications à l'analyseur 256 canaux R.C.L.²⁾ (modèle tubes à vide) qui effectue le comptage. La fonction conversion analogique digitale nécessaire pour la spectrométrie est éliminée. Les adresses binaires de la mémoire magnétique

²⁾ Voir R.C.L. 256 Channel Analyser (Argonne Type) R.C.L. Nucleonic Park - Skokie, Illinois.

sont attaquées directement en voie parallèle par les impulsions de la détection convenablement codées en système binaire. Les différentes opérations effectuées sur les signaux peuvent être suivies sur la figure 3.

3.1. AMPLIFICATION - CODAGE BINAIRE - SIGNAL SOMME

Chaque section du décacompteur attaque séparément un amplificateur transistorisé constitué par deux boucles contreréactionnées de gains 20 et 40.

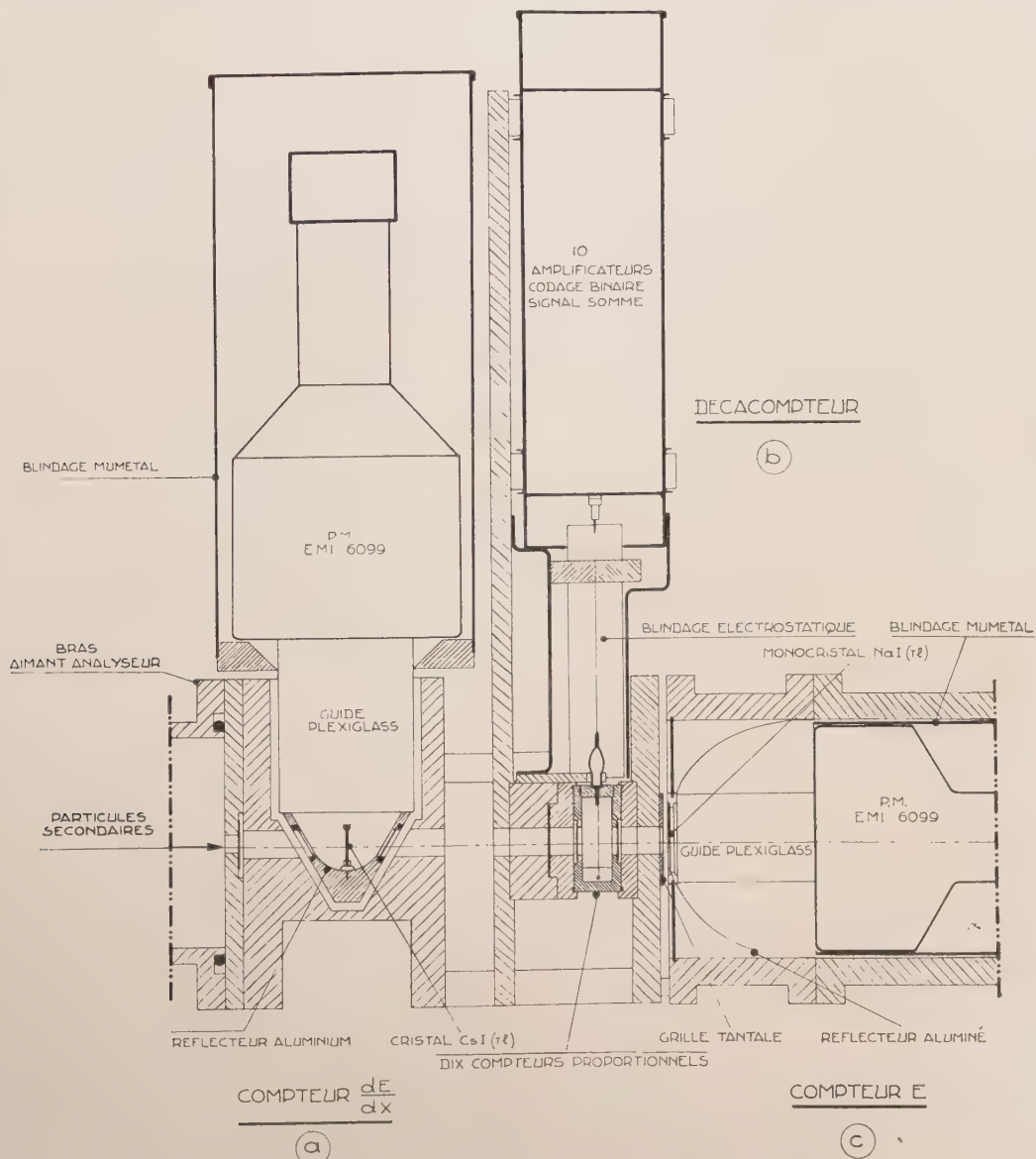


Fig. 2. Coupe de l'ensemble de détection.

La capacité d'entrée est de l'ordre de 25 pF. La boucle d'entrée est protégée par des diodes silicium contre les surtensions pouvant être produites par les compteurs proportionnels. Les impulsions du compteur sont différenciées deux fois avec une constante de temps $\tau = 3 \times 10^{-6}$ s et intégrées une

signal somme. Suivant sa valeur, on détermine si une ou deux particules sont détectées en même temps par les proportionnels. Dans la deuxième alternative le spectre est distordu car la représentation binaire est faussée. Il faut donc éliminer ces signaux par un conditionnement convenable du signal somme

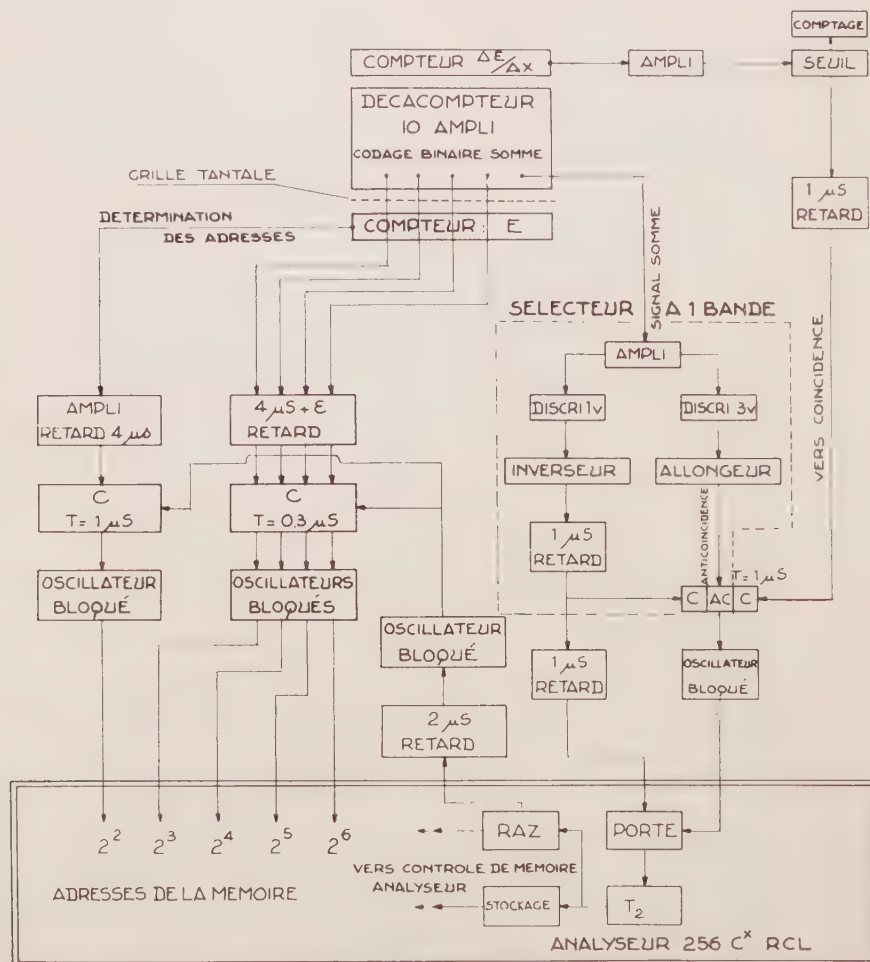


Fig. 3. Schéma fonctionnel de la détection (RAZ: Remise à zéro des adresses de l'analyseur 256 Canaux.)

fois ($\tau = 10^{-6}$ s). On obtient en sortie des impulsions de l'ordre de 4 V dont le maximum est situé à 1 μ s. Un discriminateur de seuil — 0.5 V attaqué par la deuxième boucle permet d'obtenir en sortie des impulsions d'amplitude fixe et d'une durée de l'ordre de 1.3 μ s.

Les dix impulsions sortantes sont codées par un système de diodes en quatre voies binaires pour attaquer les bascules d'adresse de l'analyseur R.C.L. Sur un point commun aux dix voies on obtient un

3.2. CONDITIONNEMENT DU SIGNAL SOMME

Les impulsions "somme" ont une amplitude de deux volts si une particule est détectée, de quatre volts si deux le sont simultanément. Une sélection de bande, constituée par deux discriminateurs à seuil réglable autour des valeurs 1 V et 3 V, permet de ne conserver que les signaux correspondant au premier cas. Le signal de sortie du sélecteur de bande peut être mis en coïncidence (temps de résolution $\tau = 1 \mu$ s) avec les signaux de la voie dE/dx

inconvénablement retardés. L'impulsion résultante commande, par l'intermédiaire d'un oscillateur bloqué, l'ouverture de la porte de l'analyseur R.C.L. avec un retard de $1 \mu\text{s}$ nécessaire pour opérer la sélection de bande. Le signal attaquant l'entrée de l'analyseur prélevé sur le discriminateur "1 V" du sélecteur de bande est retardé de $2 \mu\text{s}$ et ouvre la porte de l'analyseur ouverte si les fonctions de coïncidence sont satisfaites. Il déclenche les commandes de remise à zéro d'adresse et de stockage d'une impulsion.

3.3. ATTAQUE DES ADRESSES DE L'ANALYSEUR R.C.L.

Les impulsions des quatre voies binaires, après un retard de l'ordre de $4 \mu\text{s}$, sont mises en coïncidence (temps de résolution $0.3 \mu\text{s}$) avec le signal de remise à zéro de l'adresse retardé de $2 \mu\text{s}$, fourni par l'analyseur R.C.L. Le pouvoir de résolution de $0.3 \mu\text{s}$ a été choisi pour compléter l'élimination des codages simultanés fortuits, la sélection de bande ne pouvant protéger que des superpositions synchrones (à mieux que $0.6 \mu\text{s}$ (demi-durée d'une impulsion unique somme)). La remise à zéro étant en avance de $2 \mu\text{s}$ sur l'attaque de l'échelle d'adresse, on est certain que celle-ci est en état réceptif. L'électronique comprise entre le codage et la coïncidence ne présente pas de temps mort: en particulier les retards de $4 \mu\text{s}$ sont réalisés par ligne et non par bascule. Les bascules d'adresse de l'analyseur R.C.L. sont excitées par des oscillateurs bloqués à faible impédance de sortie.

3.4. OBTENTION DU SPECTRE À VINGT CANAUX

Le signal fourni par le compteur E est traité de

façon similaire aux quatre voies binaires décrites ci-dessus. Il est retardé de $4 \mu\text{s}$ et mis en coïncidence ($\tau = 1 \mu\text{s}$) avec l'impulsion de remise à zéro de l'analyseur R.C.L. Le signal résultant attaque, par un oscillateur bloqué, la bascule d'adresse no. 4 (2^2); les bascules correspondant aux dix sections du décacompteur sont les bascules 8 (2^3), 16 (2^4), 32 (2^5), 64 (2^6). Si le premier compteur proportionnel détecte une particule, la bascule 8 est excitée. Dans le cas où la particule est arrêtée par l'écran de tantale, il n'y a pas de signal E et l'information est inscrite dans le canal 8. Dans le cas contraire, la bascule 4 déclenche et commande l'enregistrement dans le canal 12. Chaque plage initiale de 10 mm correspondant à un compteur proportionnel est ainsi divisée en deux bandes indépendantes de 5 mm par l'intermédiaire du détecteur E. Compte tenu de cette largeur de bande, connaissant la dispersion de l'aimant analyseur sur lequel est montée la détection, on peut calculer la résolution en énergie de l'ensemble.

Dans notre cas³⁾, nous avons obtenu une résolution finale de 6 à 7×10^{-3} . La transistorisation de la plupart des fonctions fournit une excellente stabilité de marche. Les différentes bandes de détection sont absolument indépendantes et leurs réponses ne se recoupent pas sur l'analyseur R.C.L. C'est l'avantage de cette détection sur celles utilisant une localisation des particules par différence de transmission optique de la lumière⁴⁾ produite dans un cristal suivant l'endroit attaqué, procédé que nous avons utilisé il y a deux ans.

³⁾ J. Saudinos *et al.*, Comptes Rendus 252 (1961) 96.

⁴⁾ B. Sjögren, Nucl. Instr. and Meth. 7 (1960) 274.

A 200 MICROAMPERE NEGATIVE ION SOURCE, UTILISING A SUPERSONIC MERCURY JET

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Received 8 November 1960

This paper gives an account of a H^- ion source using a mercury supersonic jet type donor. The currents obtained were many times more than that given by other sources, but there are

disadvantages of this type of donor particularly when used with a vertical ion beam. It is chiefly to these disadvantages that this paper draws attention.

1. Introduction

The experiments on a mercury supersonic jet electron donor were begun in 1958, and while high H^- beam currents were obtained it became clear

that such a source was unlikely to be easily used as intended on a vertical tandem electrostatic generator. However, in the course of time it appeared that an account of the difficulties involved might be

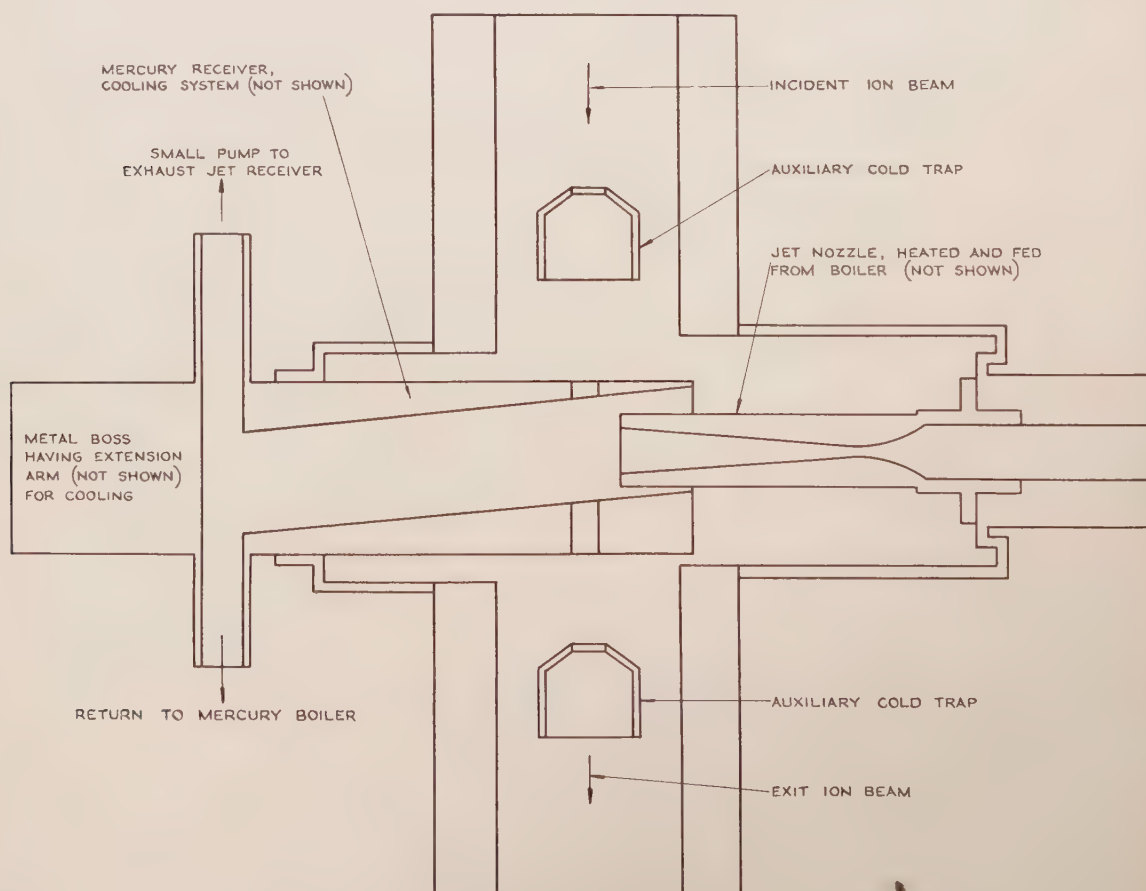


Fig. 1a. The jet assembly.

anti-bump nuclei. Violent fluctuations would occur if condensation occurred in the feed pipe.

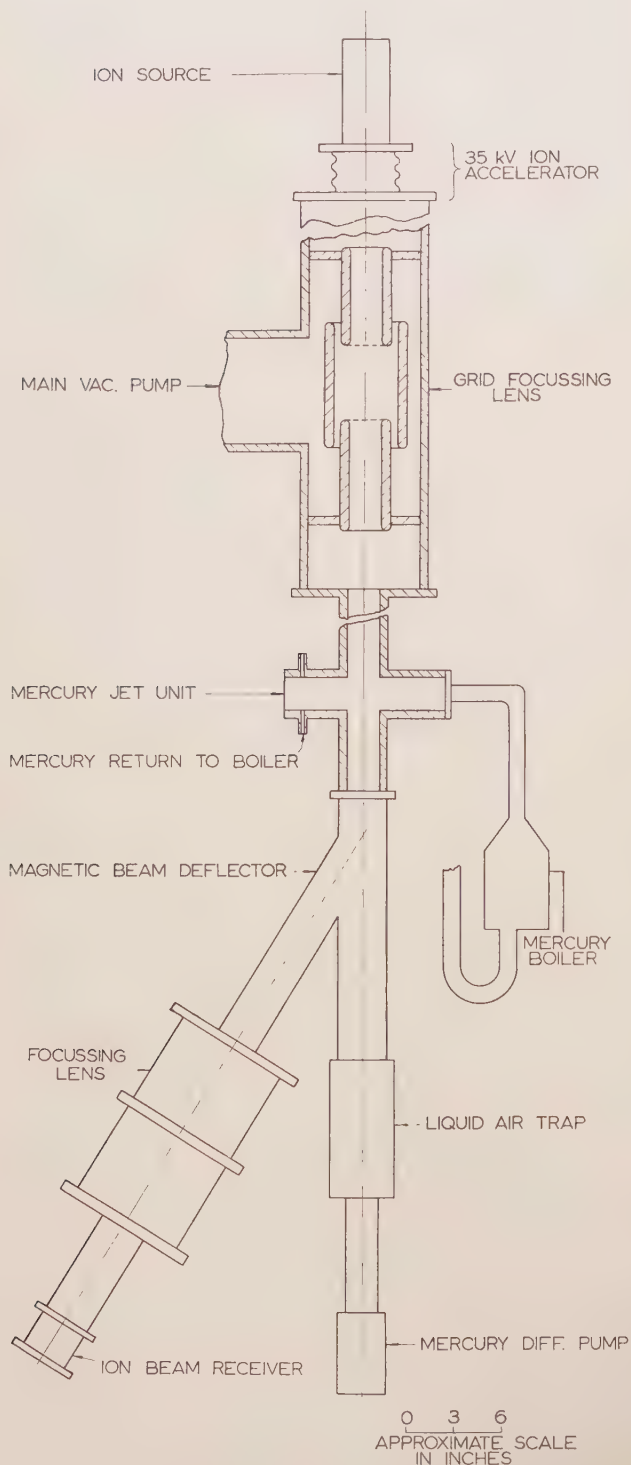


Fig. 2. The arrangement of ion source, jet unit and ion receiver.

(ii) The beam foci were diffuse at the edges. Thus the positive beam, with no jet, would focus to a spot which looked something like 5 mm in diameter, while the negative beam was more difficult to judge but could be imagined as a 5 mm core with halo fading out to 10 mm. The impairment of the optical properties of the beam is mainly due to scattering⁴) of the beam by the mercury atoms.

3.2. MERCURY THROUGHPUT

The mercury throughput was of the order of a gram per second. The boiler pressure was 3 cm and its temperature 220°C.

3.3. RECEIVER AND COOLING

The cooling of the receiver presented a problem and for any serious work the receiver would have to be thermostatically controlled at about -35°C ; that is as low as possible without freezing. In the experiments the receiver was cooled by a trichloroethylene bath to which solid CO_2 was added. Connection between the receiver and the bath was by a short 2" diameter copper rod. The trouble with this arrangement is that the cooling power, which amounts to one or two hundred watts depends directly on the mercury throughput, and if this throughput is reduced suddenly, the receiver freezes. During the freezing process the exit from the receiver is blocked and so a considerable quantity of mercury can run out of the lower beam aperture.

Besides the cooling of the receiver it is also necessary to pump it, as otherwise a back pressure is built up due to the pumping action of the jet.

3.4. AUXILIARY COOLING TRAPS

These are essential if only that they increase the H^- beam by a factor of 2. They are cooled by conduction with liquid nitrogen. The mercury which might drop from these traps on warming is no problem for the arrangements of figs. 4 and 6 but for the cases of fig. 3 and possibly also fig. 5 it would be troublesome.

The quantity is, however, small. For example after 4 hours operation the copper traps have

⁴) R. H. V. M. Dawton, E. J. M. Maunders and C. W. Trowbridge, (A.E.R.E., Harwell) to be published.

turned the colour of mercury but the deposit is very thin and would not drop off on warming. In fact, if the traps were warmed to 100°C the mercury vaporizes and is presumably taken by the pump

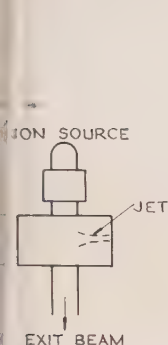


Fig. 3.

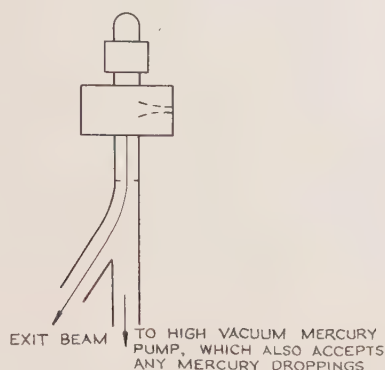


Fig. 4.

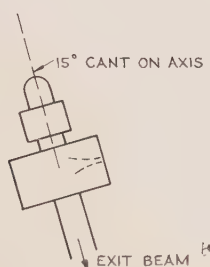


Fig. 5.

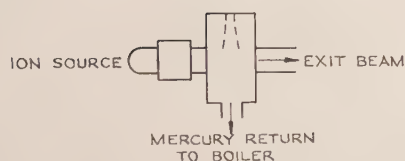


Fig. 6.

Figs. 3 to 6. Various arrangements, used for testing for mercury contamination.

or main liquid nitrogen traps. Prolonged operation of the jet could, however, result in there being sufficient mercury to drop off the traps on warming.

Beam stops must be provided to prevent trap heating by beam bombardment.

3.5. DROPPING OF MERCURY FROM THE JET UNIT

When the jet unit is used with a vertical ion beam such as in fig. 3 some mercury is periodically dropped through the ion beam aperture. Providing the jet nozzle and supply pipe were sufficiently

heated the liquid mercury drops were not from the jet itself, but from the roof of the receiver on which mercury vapour condensed and formed globules. Some of the globules would drop from the roof and make two or three bounces in the receiver before finally running out. By viewing into the ion beam aperture these bounces could be clearly seen and about once in 10 minutes a globule would bounce through the ion beam exit.

Mercury dropping also occurred by condensation on the beam aperture wall. Attempts to get this mercury away from the beam path were made by curving the aperture, fig. 7, but this was not successful as frequently the globule would detach itself from the surface and fall straight down. These mercury droppings could, however, be completely avoided by having heated inserts in the aperture. If the incident ion beam was 4 to 8 mA the halo round the beam would itself keep the aperture clear of mercury. However, if the ion beam were turned off for an hour traces of mercury could be seen building up and if the beam were then turned on a surge of pressure would go through the system.

When the equipment was working under good conditions the arrangement as in figs. 3 would drop about $\frac{1}{4}$ cc of mercury per hour, but the really objectional aspect was that the fall was in erratic quantities, about every 10 minutes. A considerable reduction, to about a tenth, of the mercury dropping



Fig. 7. Diagram of aperture used in an attempt to deflect the mercury to one side. This was not successful, as frequently the mercury would detach itself as shown with drop A.

occurred when the entire unit was tilted 15°, fig. 5, and thus reduction is believed due to the extra slope of the receiver roof causing the droplets to run towards the end of the receiver instead of dropping off.

The arrangement of fig. 4 was used for 50 to 100 hours in the laboratory and there was no evidence of mercury dropping into the exit ion beam tube. Should it be possible to use the ion beam horizontally rather than vertically then the very favourable arrangement of fig. 6 can be used.

4. Conclusion

4.1. For serious work as a negative ion source the jet receiver must be thermostatically controlled, e.g., cooling by a liquid from a temperature controlled bath. Some 100 to 200 watts must be handled.

4.2. It is inadvisable to use the mercury jet system with, e.g., an accelerator tube immediately below it (fig. 3), because from time to time a small amount of mercury will drop into the tube.

4.3. If the beam and jet are arranged as in fig. 4, the trouble mentioned in 4.2. does not occur.

4.4. From experiments using a 15° angle, fig. 5, it was concluded that an angle of 30° should be a safe minimum as this would not only reduce still further the fall through the beam aperture but suitable aperture stops should resolve completely the mercury drops and the beam.

4.5. The arrangement of fig. 6 was not tested but it will obviously cure completely the trouble with mercury droppings.

4.6. The vapour supply to the jet must be steady which means the boiler must not bump and the nozzle and feed pipe heated sufficiently to prevent condensation.

4.7. The auxiliary liquid nitrogen traps, fig. 1(a), increased the negative ion beam by a factor of 2. The disposing of the mercury which has condensed on these traps has again to be carefully considered when a vertical ion beam is used.

4.8. Beams of $200\ \mu\text{A}$ were regularly obtained. The amount of scattering of the ion beam due to the mercury target should be considered where care has to be taken with the ion optics.

Acknowledgements

The Author wishes to thank Mr. E. S. Medway for his work in assembling and testing the equipment and the Director of A.E.R.E. for permission to publish this work.

MESURE DE TEMPS COURTS PAR SEMI-CONDUCTEURS ET PHOTOMULTIPLICATEURS SATURÉS

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Reçu le 30 janvier 1961

Pulses (saturated and of definite lengths) produced on low impedances by the current of the last electrodes of a photomultiplier, are applied to semi-conductor circuits. A time defini-

tion of a few nanoseconds is obtained under normal experimental conditions.

1. Introduction

Les courants d'anode et de dynode de photomultiplicateurs de gain élevé ont été utilisés pour produire directement, dans un câble coaxial adapté au départ, des impulsions de durée définie et suffisamment saturées pour être utilisables, à la rigueur, avec un circuit de coïncidences par addition¹). Il est évidemment préférable de les utiliser avec des circuits à multiplication: les amplitudes obtenues (2 à 4 V sur 75 ohms) conviennent à l'usage de semi-conducteurs.

2. Convertisseur symétrique de temps en amplitude (fig. 1)

Chaque photomultiplicateur étant associé à un scintillateur plastique, les impulsions produites par les γ d'une source de sodium-22 et observées avec un oscilloscope Tektronix 517 A, sont reproduites sur la figure 2: les diodes d_1 et d_2 réduisent l'amplitude de la partie négative de l'impulsion.

Lorsque les diodes D_1 et D_2 sont coupées simultanément, le courant constant qui traverse la résistance r passe dans D_3 dès que le point commun aux trois diodes atteint le potentiel V_0 et va charger la capacité C , qui se décharge par la suite à travers R . L'impulsion positive de tension ainsi obtenue est transmise à un amplificateur de 2 MHz et analysée par un sélecteur à 10 canaux sous condition de la coïncidence entre deux signaux de pilotage, délivrés par deux sélecteurs à 1 canal sélectionnant une bande d'amplitude sur une dynode non saturée de chaque photomultiplicateur.

¹) R. Ballini et E. Pomelas, Nucl. Instr. and Meth. **9** (1960) 29.

Les deux bandes étant réglées vers 500 keV, la variation de l'amplitude à la sortie du convertisseur en fonction de la différence entre les deux lignes à

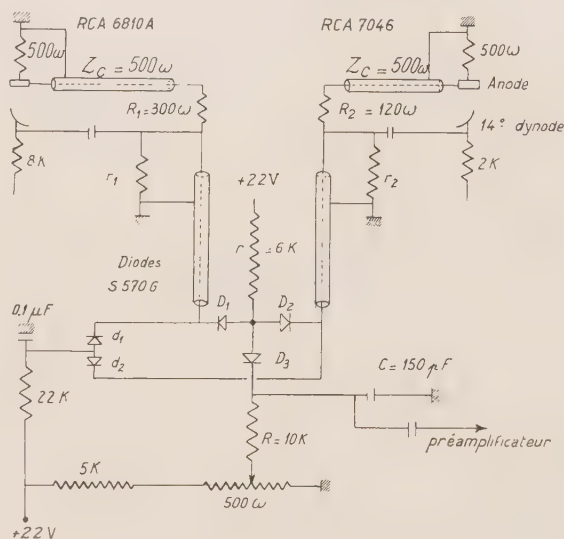


Fig. 1. Convertisseur de temps en amplitude utilisant 3 diodes à commutation rapide.

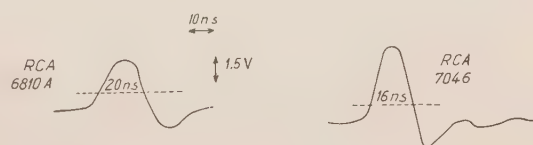


Fig. 2.

Impulsions relevées à l'entrée des diodes D_1 et D_2 (fig. 1).

retard est représentée par la figure 3a; le pointillé représente le résultat qu'on obtiendrait avec des diodes à commutation infiniment rapide et des signaux rectangulaires ayant des durées voisines de

celles des signaux réels (fig. 2) au niveau $V_0 = 1$ volt (fig. 1).

La figure 3b est obtenue en réglant entre 30 et 50 keV la bande d'énergie sélectionnée à la sortie du photomultiplicateur 6810A. Le déplacement de l'axe de symétrie est négligeable: un circuit de

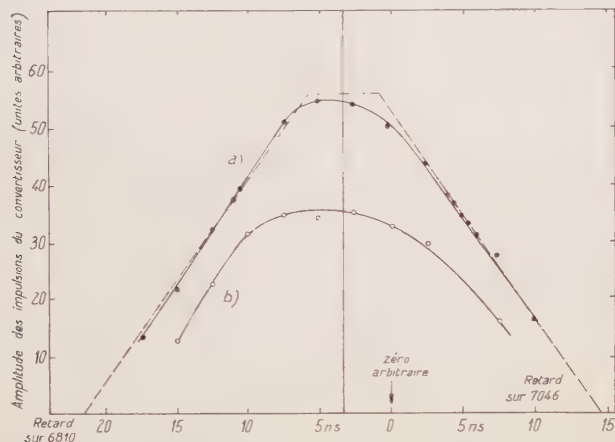


Fig. 3. a. Relation retard-amplitude pour des impulsions données par des électrons d'énergie voisine de 500 keV. Le pointillé représente la relation qui serait obtenue avec des signaux rectangulaires de 19 et 16 ns; le graphique est construit de façon à admettre le même axe de symétrie que les points expérimentaux (trait fort) et à passer au mieux par ceux-ci. b. Relation retard-amplitude utilisant des impulsions de 30 à 50 keV pour l'un des détecteurs (6810 A). Source de ^{22}Na . Scintillateurs plastiques de diamètres: 7 cm (9046) 4 cm (6810A).

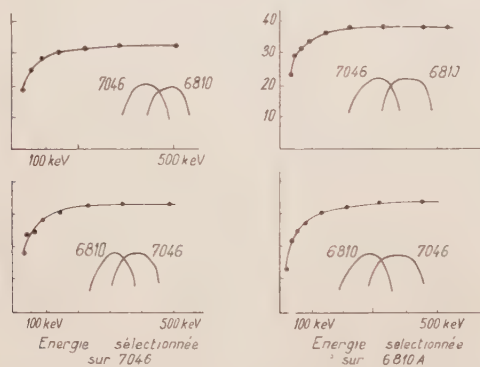


Fig. 4. Variation de l'amplitude du signal délivré par le convertisseur avec l'énergie des électrons utilisés par l'un des détecteurs.

coïncidence à 1 canal, obtenu en réglant les lignes à retard sur l'axe de symétrie et en définissant le temps de résolution par un discriminateur d'amplitudes, serait donc équilibré indépendamment de l'énergie des électrons qui fournissent l'information de temps: les sommets des impulsions observées à

l'entrée de D_1 et D_2 (fig. 1) ont, par rapport à l'instant de détection d'une particule, un retard dépendant très peu de l'énergie de cette particule.

Dans les régions linéaires, la largeur à mi-hauteur du pic est voisine de 1.5 ns pour la figure 3a et de 3 à 5 ns pour la figure 3b.

La figure 4 représente l'effet de l'énergie des électrons détectés sur la position du pic: pour chaque détecteur, deux séries de mesures concernent l'une le cas où le signal intégré sur C (fig. 1) est défini par le front avant de l'impulsion de ce détecteur, l'autre le cas où il est défini par la restitution de cette impulsion. Le déplacement du pic s'accompagne, dans tous les cas, d'une augmentation de sa largeur.

3. Autres exemples de production d'une impulsion mise en forme par débit direct du photomultiplicateur sur une basse impédance

Un photomultiplicateur EMI 9514 à 13 dynodes, associé à un cristal de NaI(Tl), a été utilisé à la place du RCA 7046 dans l'installation précédente (fig. 1): la largeur à mi-hauteur obtenue avec les impulsions de 400 à 500 keV dues à une source de sodium-22 était voisine de 2 ns avec un cristal de 25 mm de diamètre, 3 ns avec un cristal de 45 mm.

Utilisés avec des cristaux de NaI(Tl), divers photomultiplicateurs rapides de gain élevé (56AVP, RCA 6810 A et 7046) permettent d'obtenir sur un câble coaxial de 75 ou 150 ohms des impulsions de temps de montée voisin de 10 nanosecondes, durant de 20 à quelques centaines de nanosecondes, et d'amplitude suffisante (2 à 4 V) pour être appliquées directement à des circuits à semi-conducteurs.

Un scintillateur plastique ainsi utilisé ne peut évidemment donner que des signaux courts; si l'on veut éviter d'intégrer à la sortie du photomultiplicateur, des signaux longs peuvent être obtenus en utilisant une bascule rapide, telle que celle dont un usage est indiqué ci-après.

4. Convertisseur dissymétrique de temps en amplitude

L'intérêt de ce genre d'appareil^{2,3} est de distinguer entre deux retards égaux et de sens

²) R. E. Green et R. E. Bell, Nucl. Instr. 3 (1958) 127.

³) M. Bonitz et E. J. Berlovich, Nucl. Instr. and Meth. 9 (1960) 13.

avoir une grande impédance d'entrée, une lampe à commutation écran-plaque (6 BU 8) a été utilisée (fig. 7), dans des conditions où une impulsion de

[illegible]

Durée égale au retard BB'

The schematic shows a vacuum tube circuit with the following components and connections:

- Power Supply:** A $-250V$ source connected to a $1K$ resistor, which is in series with a $68K$ resistor leading to the tube's cathode.
- Grids:**
 - Control Grid (G1):** Connected to a $7K$ resistor and a $6BU8$ tube.
 - Screen Grid (G2):** Connected to a $72V$ source.
 - Suppressor Grid (G3):** Connected to a $-85V$ source.
- Plate (Anode):** Connected to a $-60V$ source and a $440\mu F$ capacitor.
- Output:** The plate is connected to a load resistor, which is in series with a $2K$ resistor leading to ground.

The graph shows the plate current I_p in milliamperes (mA) versus the plate voltage V in volts (V). The curve is a typical vacuum tube characteristic, showing a sharp increase in current as the plate voltage becomes more negative.

Plate Voltage V (V)	Plate Current I_p (mA)
-70	1.5
-72	1.4
-74	1.3
-76	1.0
-78	0.5
-80	0.1
-82	0.05
-84	0.02
-86	0.01

Fig. 7. Convertisseur de temps en amplitude.

bien à des impulsions qui se forment sur une capacité assez élevée et une résistance assez faible, permet de transmettre l'impulsion à travers un câble coaxial de 150 ohms.

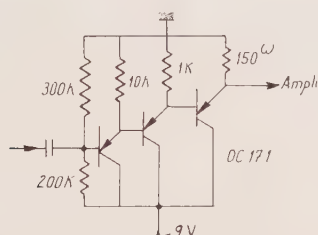


Fig. 8. Transformateur d'impédance.

La figure 9 représente la variation de l'amplitude à la sortie du convertisseur avec le retard du signal A par rapport au signal B: les deux photomultiplicateurs sont équipés de cristaux de NaI(Tl) de 2"

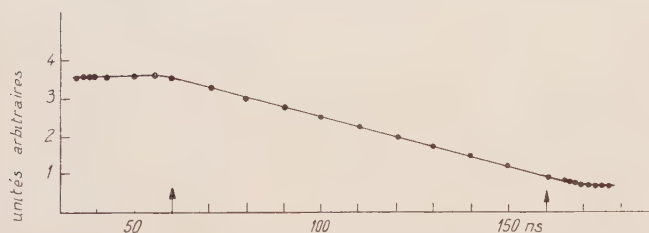


Fig. 9. Amplitude du signal délivré par le convertisseur (fig. 6 et 7) en fonction du retard apporté au signal A (repéré par rapport à un zéro arbitraire).

d'épaisseur et 1.75" de diamètre, entre lesquels est placée une source de sodium-22; toutes les impulsions délivrées par les détecteurs sont utilisées, sans sélection d'énergie par une "voie lente". La largeur à mi-hauteur est voisine de 4 nanosecondes

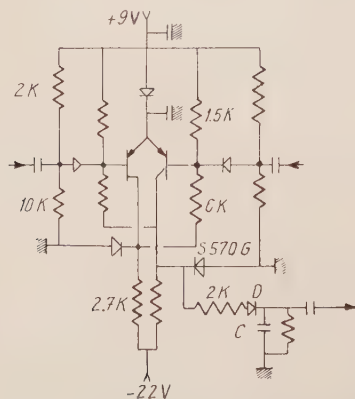


Fig. 10. Circuit remplaçant ceux des figures 6 et 7.

(3 nanosecondes lorsqu'on sélectionne les scintillations autour de 500 keV).

La région linéaire correspond à la différence des lignes de mise en forme des impulsions des deux

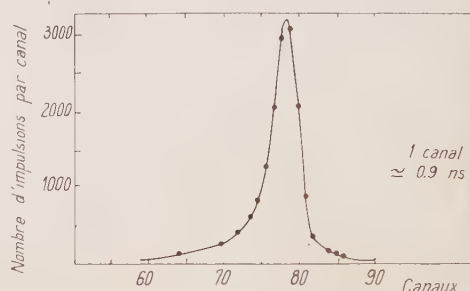


Fig. 11. Pic obtenu avec le circuit de la figure 10 sans sélection d'énergie: source de sodium-22, cristaux NaI(Tl) épais de 2" et de 1 3/4" de diamètre.

détecteurs (27 et 127 ns) c'est-à-dire au cas où une de ces impulsions est complètement encadrée par l'autre. Aux deux extrémités du graphique, le signal d'amplitude H ou h disparaît progressivement sans changer d'amplitude, pour une variation de retard qui est de l'ordre de grandeur de la définition de temps obtenue avec cet appareil: le spectre de temps est ainsi compris entre deux limites; la limite inférieure ne peut être abaissée au-dessous de 20 à 25 nanosecondes, car le signal B' doit se produire assez longtemps après la fin du signal B.

Le signal produit par la bascule est assez bien défini pour intégrer dans la capacité C (fig. 10), à travers la diode D, un courant proportionnel à l'amplitude de l'impulsion de tension sur l'un des collecteurs. Pour utiliser celui "de droite" (pas de signaux parasites), il a été nécessaire d'obtenir un

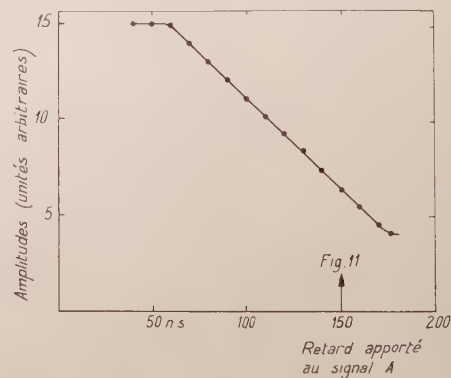


Fig. 12. Conversion de temps en amplitude par le circuit fig. 10 introduit dans le schéma fig. 6.

ront négatif plus raide, en rappelant le collecteur par une tension suffisamment négative et en définissant son potentiel minimum par une diode: ce procédé, bien que moins nécessaire avec le circuit intégrateur de la figure 7, en facilite le réglage.

La résolution obtenue avec le circuit de la figure 10 est un peu meilleure en remplaçant la diode D par deux S 570 G en série: la figure 11 montre un résultat obtenu avec ^{22}Na , sans sélection d'énergie, pour un retard de 150 ns apporté au signal A (fig. 12).

5. Convertisseur symétrique de temps en amplitude, rendu dissymétrique par un circuit de coïncidence "de supervision"^{2,3)}

Si l'on dispose d'un analyseur d'amplitudes muni d'un dispositif d'ouverture par "signal porte", ce principe peut être réalisé très simplement à partir de deux circuits analogues à celui de la figure 1, montés en parallèle mais décalés d'un retard R (fig. 13). L'amplitude du signal intégré sur la capacité C n'est enregistrée s'il y a en S un signal suffisant. Il est nécessaire d'adapter la ligne R en D₁ et la ligne de transmission T₂ à une de ses extrémités; il est prudent d'adapter la ligne de transmission T₁ au départ.

Un photomultiplicateur Radiotechnique 56 AVP (en haut sur la figure 13) et un RCA 6810 A, soumis à une tension de 2300 V, ont été utilisés avec des scintillateurs plastiques (25 mm de diamètre et

20 mm d'épaisseur) et une source de sodium-22. La largeur à mi-hauteur est voisine de 1 ns en sélectionnant les scintillations de 500 keV environ; elle est de 2.5 ns sans sélection d'énergie, le pic étant

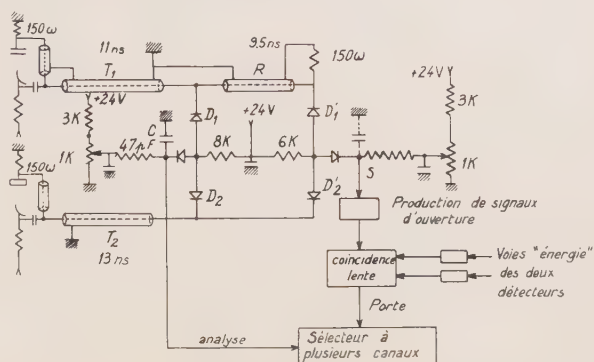


Fig. 13. Convertisseur dissymétrique de temps en amplitude utilisant un signal de coïncidence de supervision.

alors prolongé, vers les basses amplitudes, par une queue plus importante qu'avec les dispositifs utilisant une bascule. La relation entre retard et amplitude n'est linéaire que sur 5 ns en raison des courtes durées de signaux adoptées pour cet essai.

Au cours de ce travail, aucune tentative n'a été faite pour adapter parfaitement les lignes de mise en forme et de transmission, pour étudier systématiquement la forme de l'impulsion de courant à la sortie d'un photomultiplicateur et pour utiliser les particules de très faible énergie.

THE NON-PROPORTIONAL RESPONSE OF NaI(Tl) CRYSTALS TO γ -RAYS

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Measurements have been made of the variations in light output, L , with energy, E , for γ -rays which are totally absorbed in NaI(Tl) crystals. In the energy range 0.046 to 1.33 MeV the scintillation efficiency, L/E , varies by 20%, being greatest at the lowest energy. By a careful choice of calibration sources and

control of measurement conditions the results were made sufficiently precise to allow the evaluation of L/E to within 0.3% and dL/dE to within 2%. The results are used to show the variation of dL/dE with density of ionisation along the path of an electron.

1. Introduction

Thallium-activated sodium-iodide crystals are widely used for the detection of γ -rays and measurement of their energies. They find their greatest application for energies up to a few MeV, particularly for γ -rays from radioactive sources, and in this range they can provide reasonably accurate energy measurements in addition to having high detection efficiencies. In the pulse height spectrum resulting from the absorption of γ -rays in NaI(Tl), measurements are usually made from the full energy peak produced by that proportion of the γ -rays which interact and dissipate the whole of their energy within the crystal. It was long assumed that the pulse height for total absorption was proportional to the energy of γ -ray but Engelkemeir¹⁾ has pointed out that considerable variations from proportionality exist. He showed that the scintillation efficiency of a crystal, i.e. the amount of light produced per unit energy of γ -ray, varied by $\approx 15\%$ in the energy range 0-1 MeV. The efficiency was highest at approximately 50 keV and fell steadily as the energy increased. More recently Nemilov *et al.*²⁾ have found similar effects, but they noted appreciable differences between various crystal samples. CsI(Tl) and KI(Tl) were also investigated and they exhibited similar properties. Again, the variations in scintillation efficiency differed from crystal to crystal and one sample of KI(Tl) appeared to exhibit no marked deviation from proportionality.

An attempt has been made in the present experiment to measure very accurately the light output resulting from the total absorption of γ -rays in NaI(Tl) crystals. Two crystals in particular were investigated over the energy range 0.046 to 1.33 MeV. These measurements have demonstrated more precisely than previous experiments the way in which scintillation efficiency varies with photon energy, and have allowed estimates to be made of the changes in efficiency with the density of ionisation. A series of measurements of more limited scope have been made on several other crystals to compare the overall changes in scintillation efficiency.

2. Method

The two crystals upon which the more extensive measurements were made, were both cut from material provided by Hilger and Watts Ltd (Crystals No. 1 and 2 listed in the table). Each was mounted in an aluminium can with a glass end face and magnesium oxide as a light reflector. When in use the crystals were mounted centrally on a photo-multiplier, E.M.I. type 6097F, with a thin film of silicone fluid as a light pipe. After amplification, pulses from the multiplier were recorded on a multichannel analyser of the Hutchinson-Scarrott

¹⁾ D. Engelkemeir, Rev. Sci. Instr. **27** (1956) 589.

²⁾ Yu. A. Nemilov, I. I. Lomonosov, A. N. Pisarevskii, L. I. Sashin and N. D. Teterin, Izvest. Akad. Nauk. S.S.S.R., Ser. Fiz. **23** (1959) 257.

type. In order to avoid any non-linearities which could be introduced by the amplifying and recording system, calibrations were made with a high precision, high stability pulse generator. Its output was modified by an RC circuit to give the same pulse shape as that appearing at the anode of the photomultiplier, and millivolt pulses were fed in at the first valve of the amplifier. Frequent calibrations of the system were made throughout a series of measurements even though it was found that during a day the total drift of the analyser and amplifier with respect to the pulse generator was often less than 0.2%.

As photomultipliers, are well known to have high temperature coefficients, the assembly of multiplier and crystal was maintained with the temperature constant to within 0.2°C . Even so, it proved impossible to restrict the drift in output to much less than 1% in a day. To overcome this difficulty it was necessary to make frequent calibrations with a γ -ray source, for which purpose the 0.279 MeV γ -ray from Hg^{203} was used. The best results were only obtained when a calibration was made following every other measurement. Under these conditions the errors in estimation of pulse heights were in all cases likely to be less than 0.3%.

The choice of γ -ray sources depended upon several properties including availability, half life and simplicity of decay scheme. However, the primary consideration was the precision with which the γ -energies had been determined by previous workers. The most precise estimates of energy are made by crystal diffraction methods and most of the γ -rays used in the present experiment had been measured in this way. Preliminary results showed that a single measurement of energy by a magnetic spectrometer could not be regarded as reliable and when the only estimate of a γ -energy had been made in this way it had to be discarded for the present experiment. In no case was a measurement by scintillation spectrometry used to establish or confirm the energy of a γ -ray.

The range of γ -energies covered extended from 0.0465 to 1.333 MeV. An attempt was made to extend the lower limit by means of X-rays emitted from K-capture decays but it was found that the presence of K_β radiations distorted the main peak

due to K_α X-rays so that accurate measurements could not be made.

3. Results and Discussion

3.1. VARIATIONS OF SCINTILLATION EFFICIENCY

Engelkemeir found that the most illustrative way of displaying his results was to plot the scintillation efficiency against the log of energy. This method has been used in the present experiment and in fig. 1 the results for the $1\frac{1}{2} \times 1$ inch crystal are

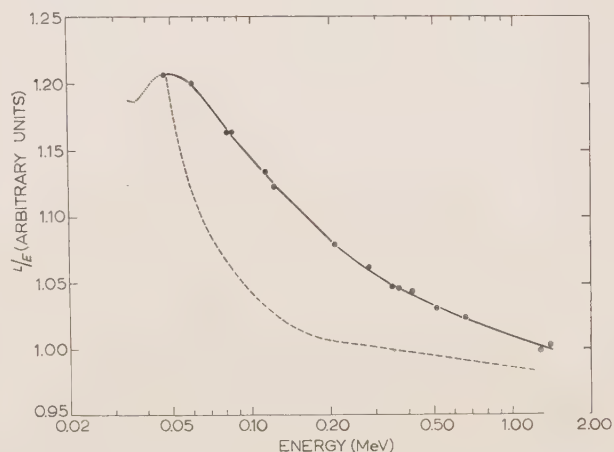


Fig. 1. L/E (full line) and dL/dE (broken line) vs energy for the $1\frac{1}{2} \times 1$ inch crystal.

shown. The dotted portion of the curve at the low energy end shows the type of variation that was indicated by the relatively inaccurate measurements on X-rays. The scintillation efficiency L/E , where L represents the light output (measured in this case as pulse height) and E the energy of γ -ray, is in arbitrary units which have been adjusted to give a value of 1 at the highest energy measured, 1.333 MeV.

Above 0.2 MeV the light output varies almost linearly with energy but the degree to which this is so cannot be demonstrated easily on a small scale graph of L against E . It is more convenient to calculate the slope of such a graph, dL/dE , using the curve in fig. 1 to smooth the data. The resulting values of dL/dE are shown by the broken line in this figure. It can be seen from the curve that between 0.2 and 1.3 MeV dL/dE varies by only 3%. In this energy region the light output is given by:

$$L = \text{constant} \times (E + 0.021 \text{ MeV}).$$

The most marked variations in dL/dE occur at energies below 0.2 MeV. In this region a very large proportion of γ -rays interacting in the crystal are absorbed by photoelectric capture in the K-shell of the iodine atom, giving rise to an electron with an energy smaller than that of the primary γ -ray by 33 keV, the K-absorption energy for iodine. The remainder of the energy is subsequently emitted as X-radiation and re-absorbed in the crystal, a

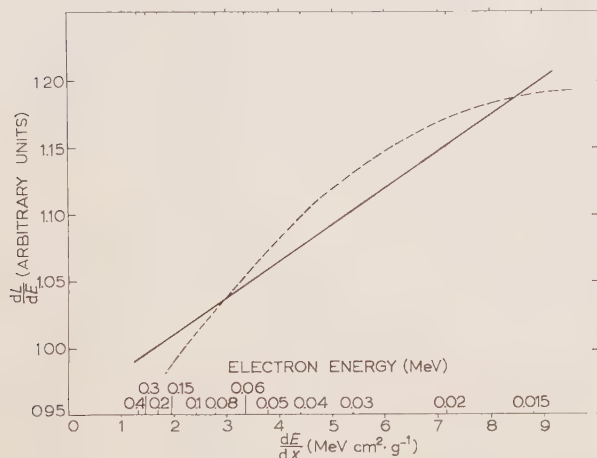


Fig. 2. dL/dE vs dE/dx for electrons in sodium iodide. The full line is for the present experiment. The broken line represents the semi-empirical theory of Meyer and Murray.

process common to all photoelectric captures whatever the γ -ray energy. When the absorption process is almost completely photoelectric a particular γ -ray energy, and hence the value of dL/dE measured in the present experiment, can be related directly to a particular energy of electron and to its appropriate rate of ionisation. This is shown in fig. 2 in which dL/dE (in arbitrary units) is plotted against dE/dx , the energy loss per unit path length for an electron in sodium iodide, the values for which are taken from the calculations of Nelms³). The total energy of an electron corresponding to a given rate of ionisation is also shown on the dE/dx axis. Errors in dL/dE are at all points likely to be less than 2%.

It is well known that the scintillation efficiency (L/E) of NaI(Tl) varies with the type of exciting particle. Ophel⁴) has found that protons produce approximately 35% more light per unit energy than fast electrons. On the other hand the more densely

ionising particles, i.e. α -particles and heavier ions, are less efficient than electrons. These changes of L/E and dL/dE with density of ionisation have been interpreted by Meyer and Murray⁵) in terms of a model which supposes that most of the light emitted by NaI(Tl) results from those excitons which are formed by the recombination of electrons and holes. After formation these diffuse until they are captured by a luminescent centre or until quenching occurs. Meyer and Murray have derived an expression based on these assumptions and fitted it to a series of experimental points representing various particles—electrons, protons, alpha particles and heavy ions of a few MeV. Part of their curve showing the variation of dL/dE with dE/dx is plotted as the broken line in fig. 2 and has been adjusted along the dL/dE axis to give the best fit. It can be seen to be in reasonable agreement with the present results.

So far the results and calculations have depended upon the set of measurements made on a $1\frac{1}{2}'' \times 1''$ crystal. A smaller crystal, $\frac{1}{2} \times \frac{3}{8}$ inch), was subjected to a similar series of measurements and the results obtained were almost exactly the same. Several other crystals were also used, all of them 2 inches long and $1\frac{3}{4}$ inches in diameter, and measurements were made for two low energy γ -rays 0.0465 MeV from RaD and 0.0596 MeV (Am²⁴¹), and two high energy ones 1.276 MeV (Na²²) and

TABLE I

Overall changes in scintillation efficiency. The amount by which the scintillation efficiency at 0.05 MeV exceeds that at ≈ 1.3 MeV.

Nr.	Crystal	Manufacturer	Increase in efficiency (%)
	(inches)		
1	$\frac{3}{8} \times \frac{1}{2}$	Hilger & Watts	17.9
2	$1\frac{1}{2} \times 1$	Hilger & Watts	20.0
3	$1\frac{3}{4} \times 2$	Hilger & Watts	17.2
4	$1\frac{3}{4} \times 2$	Hilger & Watts	18.0
5	$1\frac{3}{4} \times 2$	Hilger & Watts	20.0
6	$1\frac{3}{4} \times 2$	Harshaw	17.7

³) A. T. Nelms, N.B.S. Circular 577 (1956).

⁴) T. R. Ophel, Nucl. Instr. **3** (1958) 45.

⁵) A. Meyer and R. B. Murray, I.R.E. Trans. Nucl. Sci., N.S. 7, No. 2-3 (1960) 22.

1.333 MeV (Co^{60}). The maximum scintillation efficiency occurs in the energy region represented by the two low energy γ -rays and the two high energy ones can fairly be regarded as giving the scintillation efficiency for a minimum ionising electron. In the table the difference between the efficiency for the high and low energy γ -rays is shown as a percentage of the efficiency at the higher energy. The results are substantially similar for all six crystals but those differences that do occur must be real because errors of measurement are unlikely to amount to more than 0.5%.

The results of Nemilov *et al.*²⁾ showed that the variations in scintillation efficiency differed from crystal to crystal. In two cases they found roughly 20% changes in efficiency but in a third crystal the variations, for the energy region considered in the present experiment, may have been as low as 4%. This is the only instance in which the variations measured have been so small for a NaI(Tl) crystal. It is interesting that the results of Engelkemeir with Harshaw crystals and those of the present experiment for one Harshaw crystal and several Hilger and Watts crystals give substantially the same type of response, and that these are similar to two of the three crystals measured by Nemilov *et al.* If large differences in the variations of specific light output exist, it is surprising that most of the

results obtained are so similar. However, the aim in growing crystals is to obtain a high absolute scintillation efficiency and it is possible that this at the same time determines other characteristics. For this reason crystals chosen for scintillation spectrometry may tend to have similar degrees of non-proportionality of response.

3.2. ACCURATE MEASUREMENTS OF γ -RAY ENERGY

The attempts to measure pulse heights in the present experiment show that, provided sufficient care is taken, a single measurement of an unknown γ -ray together with accurate calibration can produce an estimate of energy with errors of less than 0.3%. This is well known to many workers, but it is worth while stating the conditions which must be fulfilled before such accuracy can be achieved. Apart from the usual requirements of stability in all the electronic equipment, the temperature of the detector must be controlled to limit the drifts in pulse height. Even so frequent calibrations with one standard radioactive source are necessary to allow for further uncontrolled drifts of the photomultiplier output. It is very important to make a critical selection of calibration sources and in addition because of the non-proportional response of the crystal, these must have energies near to that of the unknown γ -ray.

THE EFFECT OF THE NON-PROPORTIONAL RESPONSE OF NaI(Tl) CRYSTALS TO ELECTRONS UPON THE RESOLUTION FOR γ -RAYS

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This paper discusses the resolution of NaI(Tl) scintillation spectrometers for γ -rays and in particular the hitherto unexplained component usually referred to as the intrinsic crystal resolution. Experiments indicate that differences in efficiency of light collection from various parts of the crystal and variations in photocathode efficiency have only a small effect upon the resolution. It is shown that because of the non-proportional

response of NaI(Tl) to electrons, there is a contribution to the resolution arising out of the variations in number and energy of electrons involved in the total absorption of a γ -ray. Approximate calculations indicate that the magnitude of this effect is such that it could be completely responsible for the intrinsic crystal resolution.

1. Introduction

The greatest restriction in the use of NaI(Tl) scintillation counters to measure γ -ray spectra is their limited resolving power. To some extent this is due to statistical variations in the number of electrons produced at the photocathode of the multiplier, which, if normally distributed, can account for a considerable proportion of the observed resolution. However, as the efficiency of conversion of particle energy into photons and photons into photoelectrons has improved, this has become less important and the variations in light output from the crystals have become the principal limitation. This has been demonstrated by Kelley *et al.*¹⁾, who made measurements with an electronic light flasher to determine the component of the resolution due to the multiplier. They found that, for 0.66 MeV, an instrument with a resolution of 7.7% has a contribution of only 4% from the multiplier leaving unexplained a component they called the intrinsic crystal resolution, which in this case was 6.6%. (The resolution here is defined as the full width of a peak at half amplitude expressed as a percentage of the pulse height.) The intrinsic resolution of the crystal was found to vary roughly as $E^{-\frac{1}{4}}$, where E is the energy of the γ -ray.

Breitenberger²⁾ has given a general review of the factors which can affect the line width of a scintil-

lation spectrometer. From his results it can be shown that if there is a normal distribution in the number of photons produced within the crystal, the resolution, R , can be expressed as

$$R^2 = \alpha + \frac{\beta}{E} \quad (1)$$

where α and β are constants and E is the total energy dissipated within the crystal. The first term represents variations from point to point in the crystal of the transfer efficiency, i.e. efficiency with which light from an event can find its way to the photocathode and produce photoelectrons. The second term represents the contribution due to the photomultiplier statistics, both at the photocathode and in the multiplication process. Measurements of the resolution between 0.022 and 2.49 MeV made by Bisi and Zappa³⁾ show that this type of formula represents the resolution up to 0.8 MeV. When E is in MeV and R in percent $\alpha = 45$ and $\beta = 15.7$. Above 0.8 MeV significant deviations from eq. (1) occur. Bisi and Zappa assert that these are completely attributable to the way in which such γ -rays are absorbed by multiple processes. When a γ -ray is captured by a succession of events involving one

¹⁾ G. G. Kelley, P. R. Bell, R. C. Davis and N. A. Lazer, I.R.E. Trans. Nucl. Sci., N.S.3, **4** (1956) 57.

²⁾ E. Breitenburger, Prog. in Nucl. Phys. **4** (1955) 56.

³⁾ A. Bisi and L. Zappa, Nucl. Instr. **3** (1958) 17.

or more Compton encounters followed by photoelectric absorption, each individual electron sharing the energy of the original quantum is said to have a pulse height with a deviation from the mean in accordance with eq. (1). The combination of these separate effects, assuming them to be statistically independent, was found to predict the resolution at higher energies. As the experimental values of R varied as predicted in eq. (1), Bisi and Zappa suggested that the constant term from their results could be identified with α and that variations in transfer efficiency plus photomultiplier statistics would account for the observed values of the resolution.

In the experiments described below, measurements of the variations in transfer efficiency showed that they were too small to account for the intrinsic crystal resolution. Thus the assumption that the number of photons produced in an event varies normally, is incorrect. The intrinsic resolution must arise from fluctuations in the amount of light produced within the crystal. It is shown that the non-proportional response of NaI(Tl) to electrons, together with variations in the number and energy of electrons sharing the energy of the primary γ -ray, produce such an effect. The calculations described show that the non-proportional response must make a very large contribution to the intrinsic crystal resolution.

2. Experiments and Discussion of Results

The apparatus was the same as that used for the experiments described in a previous paper⁴). Careful measurements of the instrument resolution for the $1\frac{1}{2} \times 1$ inch crystal (crystal No. 1) yielded results similar to those of Kelley *et al.*¹) and Bisi and Zappa³).

Two short experiments were made in an attempt to measure the effect on the resolution of the variation in transfer efficiency. In the first a narrow collimated beam of γ -rays from Am²⁴¹, which have an energy of 0.0596 MeV, was allowed to enter the $1\frac{1}{2} \times 1$ inch crystal at various points in turn and the resulting pulse heights were carefully measured.

Such quanta are all absorbed within the first millimetre of a crystal and so, unlike those from more energetic γ -rays, all the scintillations are concentrated in a small part of the crystal. The maximum differences in pulse height noted were $\approx 2\%$.

In order to enhance the variation in light collection efficiency the above experiment was repeated with a crystal $1\frac{3}{4}$ inches in diameter and 2 inches long, which had a resolution of 8% at 0.66 MeV. The collimated beam was allowed to enter the curved face of the crystal at 12 places in turn, all approximately 0.5 inches from the face of the crystal near to the multiplier. The extreme differences in pulse height observed were 2%. The experiment was extended to cover the rest of the curved surface further from the photomultiplier and also the plane face of the crystal. Again the extreme measurements differed by only 2%.

An objection to the experiment just described is that the regions of the crystal investigated were all remote from the photocathode. The second experiment was arranged to overcome this by irradiating only the part of the crystal near to the cathode. A small piece of active indium foil producing 0.191 MeV γ -rays from In^{114m}, (which have a mean path length in sodium iodide of ≈ 1 cm) was placed at the central point between the crystal and photomultiplier, and the mean pulse height relative to that for 0.66 MeV γ -rays was measured. This was repeated with the indium source irradiating the centre of the opposite face of the crystal. The difference in pulse height in the two measurements was only 1%.

The differences in light collection efficiency with position of event in NaI(Tl) crystals have been calculated by Kuckushkin and Ratner⁵). If a crystal is considered to be transparent to its own scintillations, they estimate the overall variations in collection efficiency, for a crystal 4 cm in diameter and 6 cm long, to be 4%. This is in reasonable agreement with the present results because some of the parameters involved, for instance efficiency of the light reflector, may have differed in the two cases. When they take into account the absorption of scintillation light within the crystal, Kuckushkin and Ratner predict much larger variations in light

⁴) P. Iredale, Nucl. Instr. and Meth. **11** (1961) 336.

⁵) L. S. Kuckushkin and A. M. Ratner, Zhur. Tekh. Fiz. **28** (1958) 345.

collection efficiency than found in the present experiment. This probably means that the optical absorption coefficient used in their calculations, 0.147 cm^{-1} , does not apply to the crystal used in the experiment. The results of Van Sciver⁶⁾ show that this is probable because he finds the absorption coefficient to lie between 0.002 and 0.008 cm^{-1} .

In the theoretical derivation of eq. (1), α represents the variations from point to point in the crystal of the probability that a scintillation quantum will produce a useful photoelectron. The term takes into account all factors affecting the passage of a photon to the photomultiplier and also the variation in the photocathode efficiency. In the experiments described above, the changes in pulse height observed when various parts of the curved surface of the crystal were irradiated were probably attributable to photocathode inhomogeneities and other differences were probably due to variations in the efficiency of light collection. However, the maximum changes in pulse height measured were only 2%. Assuming a rectangular distribution of pulse heights between these limits the contribution to the resolution would be 1.3%. The experimental value determined for α by Bisi and Zappa represents a contribution of 6.7%. Clearly, neither this nor the intrinsic crystal resolution measured by Kelley *et al.*¹⁾ can be due to variations in transfer efficiency.

3. The Effect of Non-Proportional Response on Crystal Resolution

3.1. THE PROCESSES OF ABSORPTION OF γ -RAYS IN A CRYSTAL

Before discussing the effect on the resolution of the non-proportional response it will be convenient to describe the process by which a γ -ray is absorbed in a sodium iodide crystal. In the energy region considered, photoelectric absorption and Compton scattering of γ -rays are the only types of interaction that need be taken into account. Photoelectric absorption leads to the full energy of the γ -ray being expended in the crystal. In all but the very smallest crystals of sodium iodide, complete absorption can also take place by one or more Compton encounters followed by photoelectric capture. It is the number of Compton encounters

involved in the absorption of a γ -ray that is of interest in this section, and computations have been made for various energies of γ -ray.

The cross sections for the Compton and photoelectric absorption processes are well established and so the relative proportions of γ -rays undergoing these two kinds of interaction in their primary encounters were easily calculated. In following the subsequent history of a scattered quantum drastic simplifications were made. The energy range 0 to 1.1 MeV was divided into 11 intervals of 0.1 MeV and the properties of the γ -rays in a given interval were said to be represented by its mean energy. Scattered quanta from the first and all subsequent encounters were said to be emitted isotropically and their positions the crystals to be uniformly distributed over the plane through its mid-point parallel to the end faces. This approximation was necessary to allow the crystal efficiencies given by Vegors, Marsden and Heath⁷⁾ to be used to determine the probability of interaction of a scattered γ -ray. The calculations had to be made for a crystal $1\frac{1}{2}$ inches in diameter and 2 inches long because this was the smallest crystal for which suitable efficiencies were available. The energy spectrum of scattered γ -rays according to the Klein-Nishina formula was taken from the calculations of Smout⁸⁾.

Some idea of the validity of the results can be gained by comparing the peak to total ratios, which were a by-product of the calculation, with those published by Miller, Reynolds and Snow⁹⁾. For a 2×2 inch crystal and a γ -energy of 0.66 MeV, they found that 46% of all γ -rays detected fall within the full energy peak whereas the present calculation for a $1\frac{1}{2} \times 2$ inch crystal gives 49%. Considering the approximations involved this must be regarded as surprisingly good agreement.

The calculations of the following section use the distribution in the number of Compton encounters

⁶⁾ W. J. Van Sciver, Proceedings of the Large Crystal Spectrometry Symposium edited by J. G. Carver A.P.E.X.-471 p. 12 (1959).

⁷⁾ S. H. Vegors, L. L. Marsden and R. L. Heath, I.D.O. 16370 (1958).

⁸⁾ D. W. S. Smout, A.E.R.E., Report EL/R. 892 (1952).

⁹⁾ W. F. Miller, J. Reynolds and W. J. Snow, A.W.L. - 590 (1958).

taking part in the full absorption of a γ -ray. In fig. 1 this is illustrated. The graph shows the probability that, when total absorption occurs, a certain number of Compton collisions will be involved in the absorption of a 0.66 MeV γ -ray. Points representing absorption in a $1\frac{1}{2} \times 2$ inch crystal are connected by the full line. The function is of course discontinuous but the points have been connected by the line to distinguish them from others on the same graph.

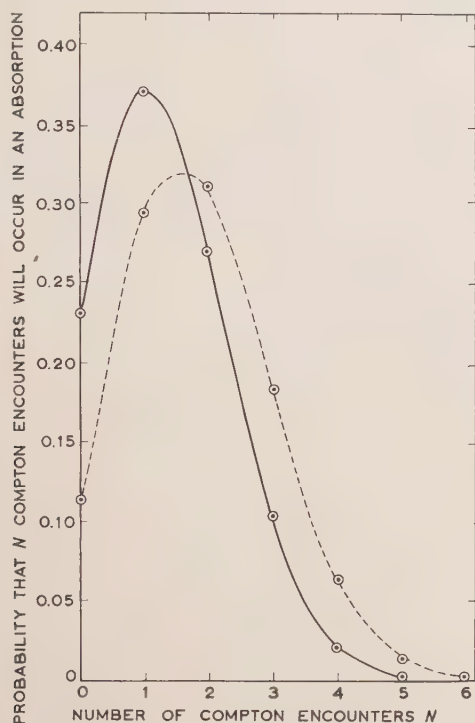


Fig. 1. The probability that N Compton collisions will occur in the full absorption of a γ -ray of 0.66 MeV. The full curve is for a $1\frac{1}{2} \times 2$ inch crystal and the dotted curve for an infinite crystal.

For comparison, calculations were carried out for an infinite crystal, and the results are shown linked by the broken line in fig. 1. It can be seen that as the size of crystal increases the distribution in number of electrons involved in an absorption does not change rapidly. For this reason it has been possible in the following approximate calculations to assume that the results for a $1\frac{1}{2} \times 2$ inch crystal can be used to represent crystals of other sizes.

3.2. THE EFFECT ON THE RESOLUTION

The scintillation light produced per unit energy loss (dL/dE) for an electron varies with the density

of ionisation, and over a wide range of energies dL/dE increases with increasing ionisation density. Thus, if the number of electrons sharing the energy of a γ -ray is increased, the total light output will also be increased because a larger proportion of the total energy is dissipated by densely ionising particles of low energy, which have a higher scintillation efficiency than the more energetic electrons. Consequently there will be a spread in light output depending on the distribution in number and energy of electrons involved in the absorption. Two processes leading to a variation are important. The first is the number of Compton encounters of the primary γ -ray, the second the number of δ -rays, i.e. electrons capable of producing further ionisation, produced in the slowing down of the electrons which originally share the γ -energy. To a first approximation the number of δ -rays produced in the absorption of a γ -ray is not dependent upon the precise method of absorption of the original quantum and so these two effects are considered separately. A further process, the production and re-absorption of Bremsstrahlung X-rays, was found to produce such a small number of additional low energy electrons that it has been ignored.

In order to simplify the calculations of the effect of the number of Compton encounters on the resolution, a convenient expression must be found giving the light produced by a single electron. The results of a previous experiment⁴) show that dL/dE for electrons is almost constant above 0.15 MeV. Thus, above this energy.

$$L = \text{constant} \times (E_e + K) \quad (2)$$

where L is the light output, E_e is the energy of the electron and K a constant which can be expressed in energy units. The value of K can be derived in the following way from previous measurements of the light produced in NaI(Tl) crystals by γ -rays and X-rays⁴).

When a γ -ray is absorbed as a result of N Compton encounters plus photoelectric capture, $N + 1$ energetic electrons are formed. Subsequent to the photoelectric capture which will almost always take place in the K-shell of the iodine atom, several low energy electrons will be produced by the emission and capture of K, L and M X-rays. Most

of the energetic electrons will obey eq. (2). Assuming that they all do their total light output will be proportional to

$$\sum E_e + (N + 1)K.$$

The amount of light added to an event by the low energy electrons produced following the de-excitation of the iodine atom will be a constant (which can be determined from measurements of the crystal response to X-rays of energy just above the K-absorption edge for iodine). The total amount of light produced by a γ -ray is given therefore by an equation of the form

$$L_\gamma = \text{constant} [\sum E_e + (N + 1)K + C] \quad (3)$$

where the last term represents the contribution of the low energy electrons. By using the average values of N computed as described in the previous section, and assuming a value for K , L/E for a γ -ray can be calculated. When K is chosen as 11.5 keV the calculated values of L/E can be fitted to the experimental curve over the range 0.25–1.05 MeV with maximum deviations of $\pm 0.1\%$. When 10.5 or 12.5 keV is chosen there are differences of $\pm 0.2\%$, representing a deviation from the experimental curve which is just significant.

In eq. (3), which represents the light produced in the absorption of a given energy of γ -ray, the only variation arises from the term $(N + 1)K$. The calculations of the preceeding section showed that N (and hence $N + 1$) has a standard deviation of 1.0 for a 0.66 MeV γ -ray. Thus there is from this source a standard deviation in the light output which is equivalent to 1.0×11.5 keV, producing a contribution to the resolution of 4.1%. It is interesting to note that for an infinite crystal the contribution becomes only 4.7%.

A further source of variance arises because an electron can dissipate its energy in different ways. It can be slowed down by directly exciting atoms within the crystal lattice or by ionisation, producing secondary electrons of energy varying from a few electron volts up to the full energy of the primary. The low energy encounters are much more numerous and represent most of the energy loss, but there are a significant number of more energetic events. For instance, an electron of 0.66 MeV produces about 3 δ -rays of energy > 10 keV and approxi-

mately 30 of > 1 keV. The luminescence efficiency for these δ -rays is different from that for the more common processes involved in the slowing down of a fast electron and because their production is a statistical process there is a resulting spread in light output.

The amount of light contributed to an event by a δ -ray of energy E keV can be determined from the results of previous measurements as the difference in response of a crystal to two energies of X- or γ -ray, one of 33 keV, the K absorption energy for iodine, and one of $33 + E$ keV. In order to calculate its effect upon the resolution it is assumed that when such a δ -ray is not produced the energy E is dissipated by processes which have a scintillation efficiency equal to that for fast electrons. The difference in light output in these two cases represents the overall effect of a δ -ray of energy E upon the total light produced by an event.

If the number of δ -rays, n , of a given energy varies normally, and if each adds to the light output of an event an extra amount of light equivalent to e keV the effect will be to produce a standard deviation of $\sqrt{n} \times e$ keV. By summing such contributions to the final variance it is found that δ -rays of energy > 1 keV add to the resolution for 0.66 MeV electrons a contribution of approximately 3.2%.

A γ -ray usually divides its energy amongst several fast secondary electrons and to estimate accurately the number of δ -rays resulting from its total absorption, the effects of the appropriate secondary electrons should be summed. However, except when the energy of a δ -ray becomes comparable with that of the primary electron, the number produced per unit energy loss of a fast electron does not vary greatly with the primary energy. To a first approximation therefore, the number of δ -rays resulting from the total absorption of a γ -ray and their effect on the resolution can be regarded as the same as that for a single electron of the same energy.

The values for the scintillation efficiency adopted for low energy electrons depended upon measurements of L/E for X-rays which were not of high accuracy. However the errors were greatest for the lower energy δ -rays, while the spread in light output arises from a small number of higher energy

nes. For these L/E was reasonably accurate, so that this source of error should not be great. No calculations have been made for δ -rays below 1 keV because their scintillation efficiency is not known. If it is similar to that for 10 keV electrons the effect of these very low energy δ -rays must be negligible. They would need to be three times more efficient at producing light than high energy electrons in order to contribute 3% to the resolution at 0.66 MeV. There is no reason to suppose that they are so ineffective and so they have been ignored.

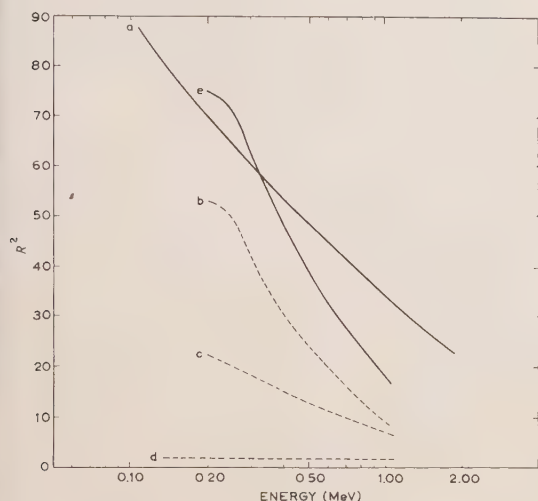


Fig. 2. The intrinsic resolution of a crystal.

The general results based on the preceding arguments are illustrated in fig. 2 where the calculated contributions to the resolution are compared with the intrinsic crystal resolution measured by Kelley *et al.*¹⁾. These results were for a 3×3 inch crystal while the present calculations involving multiple Compton encounters were for one of $1\frac{1}{2} \times 2$ inches. However, the errors introduced by comparing different sizes of crystal are not important for the present approximate calculations, especially as the number of Compton encounters in an absorption is a slowly varying function of crystal size. A graph of R^2 against energy has been chosen to present the results because the contributions from independent sources add as the sum of the squares. Curve a. is taken from the experimental results of Kelley *et al.* Curves b. and c. show the results of the non-proportional response to electrons. The former b. gives the effect of distributing the energy of the

γ -ray amongst several secondary electrons by successive Compton encounters and c. shows the contribution due to δ -rays. The variations in transfer efficiency makes a small contribution to the resolution and this is shown as the straight line d. For present purposes it has been assumed that this is not dependent on energy. Assuming that these three contributions are independent the sum of their effects is represented by the curve e. This has not been continued below 0.2 MeV because as the energy decreases below this value the assumptions made become increasingly invalid.

It can be seen that the calculated values for the intrinsic resolution are of the same order as the experimental ones and show a similar decrease with increasing energy. At all energies between 0.2 and 1.0 MeV the calculated resolution is within 30% of the experimental value. This good agreement indicates that the non-proportional response of the crystal to electrons accounts for a very large portion of the intrinsic crystal resolution.

4. Summary and Discussion

The results of the calculations have shown that the non-proportional response of NaI(Tl) to electrons makes a very large contribution to the crystal resolution for γ -rays, owing to the variations in the way the γ -ray energy is distributed amongst the electrons which finally dissipate its energy within the crystal. It is possible that this effect could be completely responsible for the intrinsic crystal resolution. However, the present results are of an approximate nature, particularly so for δ -rays, and further more accurate calculations are needed. Before this is done it is important that the scintillation efficiency for low energy electrons should be determined. This may be done by bombarding the crystal with electrons of the appropriate energies or possibly by using a similar method to that used here but with mono-energetic X-rays selected by a crystal spectrometer.

An experimental check on the contribution of the non-proportional response to the intrinsic resolution could be attempted if measurements could be made on crystals with varying degrees of non-proportionality. However, it may be difficult, or even impossible to grow crystals with a marked

difference to those used in the present experiment, especially as a high scintillation efficiency is an added requirement. A second approach which may prove more profitable is indicated by the work of der Mateoson *et al.*¹⁰). With crystals grown by the Kyropoulos method they found that the ratio of the scintillation efficiency for alpha particles to that for electrons varied with temperature and as the crystal was cooled this ratio approached unity. It was also noted that the resolution varied with temperature but that the changes were not closely dependent on the light output. One crystal had its best resolution for α -particles at -175°C when its

¹⁰) E. der Mateoson, M. McKeown and C. O. Muelhause, *Phys. Rev.* **103** (1956) 967.

light emission was least. It is possible that some of these variations could be due to effects of the type discussed above. For instance, statistical variations in the number of δ -rays formed by the parent α -particle could have produced a spread in light output which disappeared when the non-linearity was removed. Measurements of the intrinsic resolution and non-proportionality for α or γ -rays at various temperatures might therefore give some estimate of the contribution to the resolution made by variations in the scintillation efficiency.

I would like to thank Mr. R. B. Owen for many useful discussions and Mr. G. W. Hinder for his help with the experimental work.

A MAGNETIC SPECTROGRAPH FOR ANGULAR DISTRIBUTION MEASUREMENTS

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The design and performance of a high-resolution, broad range magnetic spectrograph are described. The angle of entry at the pole edge is chosen to give second order focussing, so the recorded line width is almost entirely due to the width of the object slit and target thickness. The instrument has an energy resolution

of 1040, using a natural α -emitting source and is thus suitable for studying the properties of closely-spaced nuclear energy levels. An energy range of three to one is recorded on a single plate for a given field and six exposures may be carried out in succession without breaking the vacuum.

1. Introduction

The present interest in nuclear spectroscopy has led to the design of many instruments using magnetic fields to analyse the momenta of charged reaction products from the ion bombardment of thin targets. When investigating groups of particles corresponding to closely spaced nuclear levels, a

high resolution is required both in the analysing system and beam energy selector.

The ion beam accelerated by the A.E.I. electrostatic generator¹⁾ and analysed by the electrostatic energy selector²⁾, has an energy resolution of 1200 or better. The resolution of the reaction analyser was chosen to be approximately 1000.

† Now at the Rice Institute, Houston, Texas.

¹⁾ D. R. Chick and D. P. R. Petrie, Proc. I.E.E. **103B** (1956) 132.

²⁾ S. E. Hunt, D. P. R. Petrie, K. Firth and A. J. Trott, Proc. I.E.E. **103B** (1956) 146.

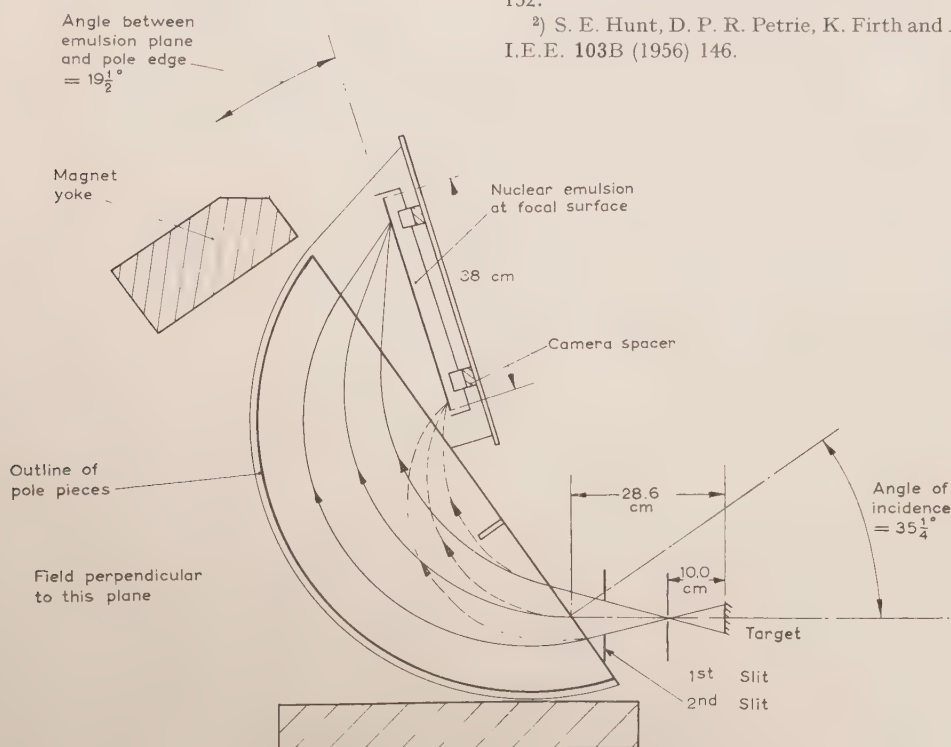


Fig. 1. Schematic diagram of magnetic spectrograph.

When a large number of particle groups covering a wide range of energies is of interest, as in the present case, it is desirable to have reaction products with a broad range of energies, simultaneously recorded. Two types of broad range magnetic spectrograph have been used for the measurement of the angular distributions of closely spaced groups from nuclear reactions. The first type, in which the particles are analysed in a uniform magnetic field with circular pole edges, has been described³⁾. The second type, of which the present instrument is an example, was proposed by P. H. Fowler of Bristol. It uses a uniform magnetic field, with a straight pole edge. Two instruments of this type have been described^{4,5)}. Only the first of these could be used for angular distribution measurements. In each case the maximum radius was about 20 cm and the resolution attained was considerably lower than in the present instrument.

In the straight-pole-edge, uniform-field instrument second-order focussing is achieved when the angle of incidence is $\sin^{-1}(1/\sqrt{3})$. This means that the first and second derivatives of the co-ordinates of the focus, with respect to the angle of incidence vanish and the aberration width is then proportional to the cube of the angle of acceptance in the median plane. Fig. 1 shows the particle trajectories in the instrument. The narrow first slit forms the object and after deflection through 110° the particles are focused on a surface which approximates closely to a plane inclined at $19^\circ 29'$ to the pole edge. Since the centre of curvature lies on the opposite side of the normal to the pole edge, from the object point and image point, the solid angle is reduced by a factor of about 2 due to the Z-defocussing effect perpendicular to the median plane which affects both the entrance and exit rays.

The maximum proton energy was chosen to be 15 MeV, which for a maximum measurable magnetic field of about 14.5 kG corresponded to a radius of curvature of about 39 cm. The ratio of the extreme particle energies recorded on a 15 in. nuclear plate placed in the focal plane is approxi-

mately 3 for particles of the same charge-to-mass ratio.

The instrument is mounted on a turntable to enable angular distributions to be determined, the target centre being made to coincide with the axis of rotation of the turntable. A photographic plate held at a fixed angle to the beam direction is used to monitor the reaction when the target is unstable under irradiation, thus enabling spectrograph exposures taken at different angles to be correlated.

2. Theory

The theory of the focussing properties of the spectrograph neglecting fringing field effects is as follows.

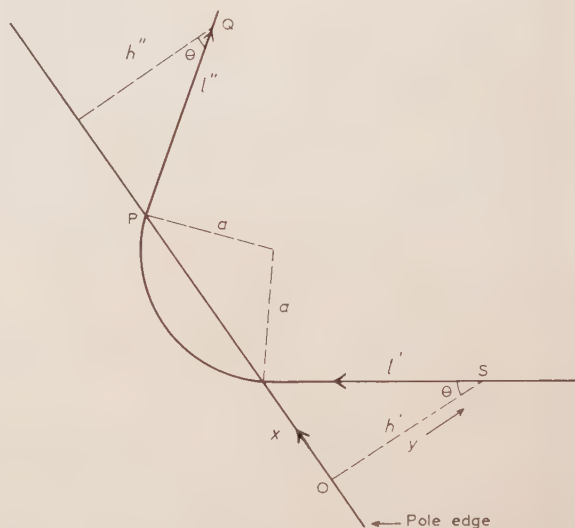


Fig. 2. Diagram showing quantities used in focussing theory.

The co-ordinate system in the median plane is shown in fig. 2. S is the intersection of this plane and a line source perpendicular to it. The equation of the emergent ray PQ is:

$$y = mx + c \quad (1)$$

where

$$m = \cot \theta \quad \text{and} \quad c = -(2a \cos \theta + l' \sin \theta) \cot \theta.$$

If θ is increased to $\theta + d\theta$

$$y + \delta y = (mx + c) + (\dot{m}x + \dot{c})\delta\theta + (\ddot{m}x + \ddot{c})(\delta\theta)^2/2 + (\ddot{m}x + \ddot{c})(\delta\theta)^3/6 \quad (2)$$

where the dotted quantities are derivatives with respect to θ .

³⁾ C. P. Browne and W. W. Buechner, *Rev. Sci. Instr.* **27** (1956) 899.

⁴⁾ D. R. Inglis, *Phys. Rev.* **78** (1950) 104.

⁵⁾ D. J. Prowse and W. M. Gibson, *J. Sci. Instr.* **33** (1956) 129.

First order focussing occurs at a point $(x_1 y_1)$ such that

$$\dot{m}x_1 + \dot{c} = 0.$$

By differentiating m and c as defined above

$$y_1 = 2a \cos^2 \theta \sin \theta - l' \cos \theta$$

$$\text{and} \quad x_1 = 2a \cos \theta (1 + \sin^2 \theta). \quad (3)$$

If the radius of curvature, a , is eliminated between x_1 and y_1 the locus of foci is found to be the straight line

$$y_1 = \frac{\cos \theta \sin \theta}{1 + \sin^2 \theta} x_1 - l' \cos \theta. \quad (4)$$

Second order focussing occurs if $\dot{m}x + \dot{c} = 0$. After differentiating m and c twice, this reduces to the condition that

$$\theta = \pm 35\frac{1}{4}^\circ \quad (5)$$

but only the positive value is applicable.

If the value in (5) is adopted, the third order terms in (2) give an aberration

$$\delta y = -1.63a (\delta\theta)^3 \quad (6)$$

and also (4) represents a line at $19\frac{1}{2}^\circ$ to the x -axis. The magnification of the instrument measured perpendicular to the rays is found to be unity. If distances along the focal line are denoted by Δs it can be shown from (3) and (4) that the relationship between Δs and a change Δa is

$$\Delta s = \frac{4}{\sqrt{3}} \Delta a. \quad (7)$$

Whence the dispersion of the system is:

$$\frac{\Delta s}{\Delta E/E} = \frac{2a}{\sqrt{3}}. \quad (8)$$

It may be shown from the geometry of the system that for a small variation $\delta\theta$ in θ , causing a change δy in y , the distance along the line of foci

$$\Delta s = \delta y. \quad (9)$$

From (6), (8) and (9) the aberration width for an angular spread $-0.03 \leq \delta\theta \leq 0.03$ radian is

$$\delta y = 2 \times 1.63 \times (0.03)^3 a$$

$$\Delta s = 9a \times 10^{-5}$$

$$\text{or} \quad \Delta E = 7.7 \times 10^{-5} E \quad (10)$$

or an energy resolution of 13 000, which is the theoretical limit for a line source with this angular aperture, neglecting fringing field effects.

The effect of a source of finite area can be estimated easily by displacing the line source perpendicular to the incident ray, for example by 0.2 mm. Then the displacement on the line of (4) is $\Delta s = 0.2 \operatorname{cosec} \theta = 0.34$ mm. This is much larger than the aberration width of 0.04 mm for the maximum radius of ≈ 40 cm.

3. Requirements for Angular Distribution and Q-Value Measurements

3.1. TARGET SYSTEM

The system used consists of a large target illuminating a small object slit (fig. 1), in preference to the alternative of replacing the object slit by a target of the same size. This choice was determined, in the case of the present instrument and beam arrangement, by the backward limit of angles of observation in the two cases. The object slit of the present spectrograph must be within 7 in. of the pole edge if the full length of the 15 in. detector is to be used. With the target at this position the largest angle of observation with respect to the beam would be 90° , whereas with the present system the maximum backward angle is 115° .

If the beam density is the same in the two cases, the numbers of particles detected from a uniform target layer are equal, provided that the target in the present system is large enough to fill the same acceptance aperture in the median plane permitted by the aberrations of the instrument. It was considered that the majority of the available beam could be concentrated on a target whose size is such that about 1 in 2.5 of the maximum aperture was filled, and so no appreciable loss in collecting power would occur due to this smaller target size. There is, however, some loss in collecting power caused by shadowing of part of the target area by a spacer between the pole faces. This is discussed in section 5.

The variation of the solid angle of collection at different points of the target for both the spectrograph and monitor system must be considered in the present arrangement. For a large target this variation may be considerable, thus making symmetry of emission about the target centre desirable.

3.2. Q -VALUE MEASUREMENT AND ENERGY CALIBRATION

The accuracy with which a reaction Q -value may be determined depends upon the measurement of the energy of the bombarding and emitted particles, the angle between these particles and the masses of the nuclei partaking in the reaction. This is true for the more precise relativistic equation as well as for the non-relativistic case.

The energies of the bombarding and reaction product particles are measured from their deflection in the magnetic spectrograph which was calibrated as described in section 5. The reproducibility of this calibration depends on the relative locations of the entrance slit of the spectrograph, the pole edge and the detector. The magnetic field at any one point of the particle trajectory should have a constant ratio to that at the reference point, as the field strength is changed. This linear relation is expected to break down in the fringing field region as saturation of the pole pieces is approached so that the calibration should be carried out for the full range of magnetic fields to be used.

The target and first slit are located reproducibly with respect to the pole edge and detector by means of dowels. The different points of the nuclear emulsion plate used as detector are related to the pole edge by a reference system of dots printed by exposure to light through an array of holes drilled in the plate holder. The accuracy of reproducibility required to measure the energies of emitted particles to 1 part in 1000 is 0.005 in. overall. A more detailed account of the mechanical system ensuring this is given in section 4.

To calibrate the instrument, natural α sources having known momenta have been used (section 5). These calibration lines have been related to the reference dot system using an arbitrary criterion for line position. This was taken to be the intercept of the high energy edge of the line on the momentum axis.

3.3. ANGULAR DISTRIBUTION MEASUREMENT

The relative differential cross-section at an angle θ for a reaction is proportional to the product $(N_0/N_1N_2)\Delta\Omega$, where N_0 is the number of particles counted in the detector, N_1 the number of nuclei

in the irradiated target, N_2 the number of nuclei/cm² in the incident beam, and $\Delta\Omega$ is the solid angle of collection at the angle θ . Since, with the magnetic spectrograph, exposures at only one angle at a time are possible, the factors N_1 , N_2 and $\Delta\Omega$ should remain constant or vary in a known manner for the series of observations. To ensure adequate knowledge of N_1 and N_2 some system of monitoring is necessary to relate exposures at different angles. The incoming beam of particles is collected and the beam current integrated to give a measure of the total charge falling on the target. This method does not detect deterioration in the target resulting in changes in N_1 .

A method of reaction monitoring, in which a nuclear emulsion is held at a fixed position relative to the target to detect reaction products from target nuclei, has been used. This measures changes in the relative values of the product N_1N_2 for each of the irradiations. The solid angle of collection $\Delta\Omega$ must remain constant over the full range of angles of observation. This requires that the target centre should lie on the turntable axis. This axis is arranged to be parallel to the median plane of the magnet and the perpendicular distance between it and the median plane is small compared to the pole gap. The system designed to satisfy these requirements will be described in section 4.

The solid angle will also vary over the limits of the detector. This variation should be known if relative angular distributions are to be measured, since the emergent energy of particles from a nuclear reaction varies with angle of observation, thus changing the line position on the plate. This variation has been calculated and also checked experimentally (section 5).

4. Design

4.1. MAGNET

The magnet rests on four lifting jacks standing on a steel plate supported by a large, 67 in. diameter ball race, whose fitting tolerance is 0.004 in.

The turntable is geared by a rack and pinion through reduction gears to a manually operated handle by which the angle of the turntable may be set to an accuracy of 5 min of arc.

The current windings of the magnet are of

aluminium of $\frac{1}{2}$ in. square cross-section with a $\frac{5}{16}$ in. diameter hole through which cooling water passes. The rate of flow of cooling water is about 5 gal/min which, combined with a cooling fan in the pumping circuit, keeps the magnet temperature constant during a single day to within one or two degrees.

The magnet yoke, pole limb and pole pieces are made from low carbon mild steel. The parallel pole pieces, which are 2.5 in. thick to withstand magnetic forces, are held to the pole limbs which are fixed to the yoke. Since the uniformity of the field affects the resolution which may be attained with the instrument, the pole pieces are machined flat, the gap of 1.5 cm being constant to 0.1%. The pole pieces form the sides of the vacuum box which is completed by an arc of brass cemented to them.

4.2. FIELD SUPPLY AND MEASUREMENT

The exciter current for the coils is supplied by a D.C. generator, the regulation and current stabilisation of which have been described previously⁶). The requirement for maximum resolution and Q -value measurement was that the field should be accurately reproducible to 1 part in 10^4 . The field is stabilised using the signal from a proton resonance probe to regulate the magnet supply current. It has been found experimentally that the field drift contributes negligibly to the line width. Field stabilisation is possible over a continuous range of fields using a frequency meter as reference, or at six fields corresponding to six crystal-controlled frequencies.

The field measurement at the reference point is accurate to 1 part in 10^4 , its degree of reproducibility being the same.

Hysteresis appears to have a negligible effect upon the trajectory for a given measured field.

4.3. Q -VALUE MEASUREMENT

The accuracy with which the Q -values of a reaction may be determined depends upon the accuracy of energy and angle measurement. Since the instrument is not designed to be absolute, it must be calibrated initially. Further measurements of energy depend upon the geometric and magnetic reproducibility of the system. It is, therefore,

⁶) R. Bailey and E. C. Fellows, J. Brit. I.R.E. **19** (1959) 309.

necessary that the slit and camera be accurately located relative to the pole edge.

The location of the first slit has been ensured by a series of dowels and spigots which locate it relative to the pole edge and perpendicular to the median plane.

The momentum calibration is made relative to a reference system of dots printed on the nuclear emulsion by illuminating a series of holes, spaced equidistantly at 1 cm, which are drilled in the centre turret of the camera. Six 15×1 in. nuclear plates, which may be exposed successively without breaking the vacuum, are held fixed in a hexagonal turret in the camera by means of springs acting in three perpendicular directions. The whole plate turret is dowelled to the base plate which is in turn positioned relative to the pole edge by means of dowels.

The angular setting of the magnet relative to the beam direction is known to about 0.0015 radian. (This uncertainty corresponds to an error of emission of less than 1 part in 2000 for the worst case.)

4.4. ANGULAR DISTRIBUTION MEASUREMENT

4.4.1. Target Chamber

The target chamber, which is coupled to the beam tube by flexible bellows, has three exit ports. The angular range on each port is $\pm 22\frac{1}{2}^\circ$. The total angular range is $0^\circ_{\text{Lab}} - 115^\circ_{\text{Lab}}$, the backward angular limit being set by the size of the magnet coils and beam tube and the distance between the target and object slit. The target is held fixed relative to the beam direction, being mounted from a large diameter thrust race which enables the target chamber to be moved while the target is held at a fixed angle of $\pm 45^\circ$ relative to the incoming beam.

The exit flanges on the target chamber are retractable so that the magnet may be rotated over the total angular range without altering the position of the geometric target centre.

4.4.2. Monitoring

The beam charge is collected on a copper strip which is placed immediately behind the target position, so that it covers the total angular range of 45° on each port.

A nuclear emulsion plate is mounted at a fixed angle of approximately 75° to the beam direction, to monitor a reaction product characteristic of the nucleus to be studied. The target is inclined at 20° to the vertical so that its mount does not shadow

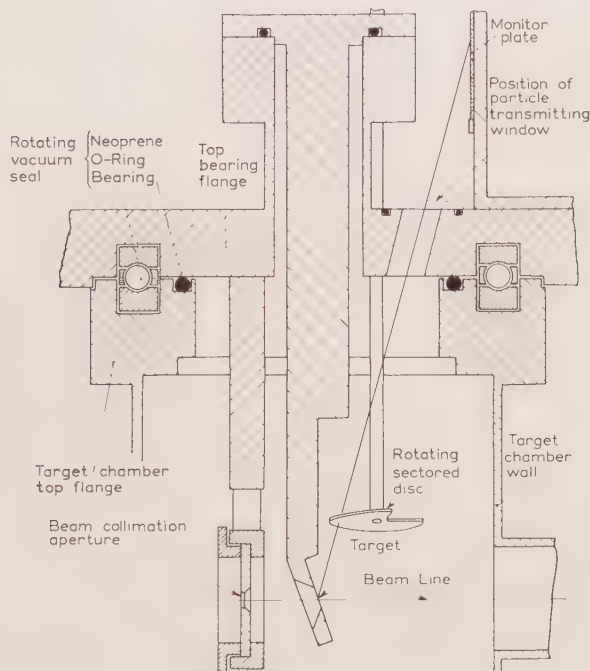


Fig. 3. Target mounting showing monitor plate (vertical section).

the portion of monitor plate used (fig. 3). A reference system of dots is printed on the emulsion by illumination through the plate mounting. The geometric relation between this system and the target centre is known accurately and hence the absolute solid angle subtended by unit area of the plate at the target is known.

4.4.3. Target Illumination

The irradiated target area is a circle of 6 mm diameter centred on the geometrical target centre which lies on the axis of the slit system. The circular area is defined by an aperture which is elliptical since the target plane is inclined to the beam direction. This aperture is held in a mount dowelled to the flange holding the target and monitor plate. The integrated beam density falling on the target must be uniform. Large deviations

from uniformity can be detected from the observation of target marking after exposure.

The vertical ion beam from the electrostatic generator is deflected through 63° by an electrostatic energy selector²⁾. The beam is bent through a further 27° by a bending magnet. A quadrupole magnetic lens is placed between these two components to focus the beam in the horizontal plane on the target, which is at a distance of about 8 m. A slit is inserted in the beam tube between the target and the 27° bending magnet. An error signal, fed from the jaws of this slit to the exciter current of the bending magnet, reduces fluctuations in intensity of target illumination due to beam position instability⁷⁾.

5. Tests and Calibration

5.1. FOCAL POSITION

The position of the plate holder for optimum focussing was determined by finding the minimum width of a line, from an alpha-emitting source, on the plate. The source used was $\text{Po}^{212}(\text{ThC}')$ which was prepared from an emanating Th^{238} liquid source. The minimum source thickness obtained was 4 keV (section 5.3). By repeated exposures of this source using three different fields while the plate was moved parallel to itself, an optimum position was observed for three positions along the plate. The angle of incidence used in these tests was approximately equal to that calculated for second order focussing neglecting fringing field effects (section 2).

Calculations on the effect of fringing field upon the optimum angle of incidence have shown that it is negligible.

The focal plane was found to be inclined at 20° to the pole edge instead of the $19\frac{1}{2}^\circ$ predicted by simple theory neglecting fringing field effects (section 2). Fringing fields do not, however, change this angle so this discrepancy is unexplained.

5.2. DISPERSION AND MOMENTUM CALIBRATION

The instrument has been momentum calibrated using α -emitting sources. A source of Am^{241} was used to obtain the dispersion by varying the field

⁷⁾ D. G. Keith-Walker, A.E.I. Research Report A 1020 (1960).

setting, the energy of the emitted α -particles remaining constant. It is necessary in such measurements to assume that there is no saturation in the pole pieces and hence that the radius of curvature may be expressed as a function of distance along the plate independently of field. Since the momentum of the α -particles from Am^{241} is not a widely accepted standard, the α -particles from ThC and ThC' were used as absolute standards of momentum. Assuming a linear calibration, the relative determinations of radius from Am^{241} give, for a slightly modified target and slit assembly, a value for the line position versus radius of $\Delta s = (-2.310 \pm 0.003)\delta\rho$, where Δs and $\delta\rho$ are, respectively, the distance along the plate and the corresponding change in radius. The dispersion of the instrument may be calculated from this relation to be $-(1.155 \pm 0.0015)\rho/100 \text{ cm}$ for 1% change in energy, compared with the theoretical value of $1.1546\rho/100 \text{ cm}$ for 1% change in energy.

The reproducibility of line position, allowing for resetting accuracy of the magnetic field and reproducibility of camera position, is about 1 part in 4000 in momentum.

5.3. LINE SHAPE

The total line width from a reaction is composed of contributions due to the width of the first slit, instrumental aberrations, angular spread perpendicular to the median plane and finite target thickness. The contribution to total line width due to instrumental aberrations, for trajectories both on and off the median plane, has been shown to be negligible. The theoretical line shape for a first slit width of 203μ is almost rectangular with a full width at half maximum of 345μ . This is less than the observed width at half maximum of the 8.78 MeV α line from Po^{212} which was 380μ . The difference is attributed almost completely to source thickness which is thus calculated to be about 4 keV full width at half maximum. The resolution $E/\Delta E$, where ΔE is the full width at half maximum, is in this case about 1040, the source width being included in this figure.

5.4. DISTRIBUTION FROM AN ISOTROPICALLY EMITTING SOURCE

The angular distribution from an Am^{241} source

has been measured for a single value of magnetic field. When the instrument is properly adjusted, such a distribution is expected to be isotropic. The angular distribution obtained experimentally confirmed this assumption the standard deviation from the mean being $\pm \frac{1}{2}\%$, the error on each point being $1\frac{1}{2}\%$ due to counting statistics. The instrument is thus not expected to contribute any significant distortion to an angular distribution.

5.5. VARIATION OF SOLID ANGLE ALONG THE DETECTOR PLATE

The relative variation in the solid angle of collection subtended at the centre of the target for a first slit of 0.2 mm width, as a function of the distance from the high energy end of the detection plate is

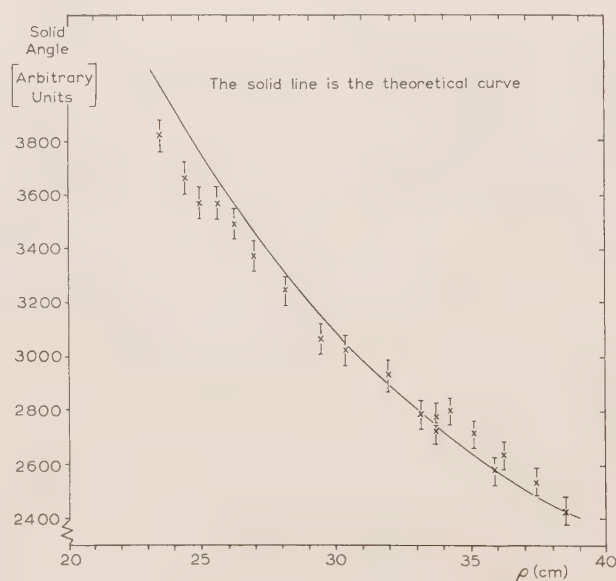


Fig. 4. Relative variation of solid angle as a function of radius.

shown in fig. 4. This variation agrees reasonably with the shape of the curve calculated from the theory allowing for the Z -defocussing effects⁸). The experimental points have been corrected for shadowing by the spacer between the pole faces, mentioned in section 3.1. This correction amounts to approximately 6% at the top of the plate and 30% at the bottom.

The theoretical and corrected experimental values of the absolute cross-section at a radius of

⁸) W. Cross, Rev. Sci. Instr. 22 (1951) 717.

33.7 cm for an 0.6 mm slit agree, their value being 4.4×10^{-5} steradians.

For comparison with other instruments, the solid angle subtended by the instrument at the first slit may be more appropriate. Its calculated value in this case is about 5×10^{-4} steradians.

5.6. CONCLUSION

The instrument has been used with a beam of deuterons from the 4 MeV A.E.I. electrostatic generator to study the properties of closely spaced nuclear energy levels excited by (d, α) and (d,p) reactions. In such use, the resolution has been approximately 700 for 5 MeV protons, the decrease from the figure of 1040 quoted in source work being due to target and backing thickness, and beam energy spread.

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The design and construction of the spectrograph has been shared by the design drawing offices and workshops of this Laboratory and A.E.I. Manchester. The magnetic field stabilisation system is due to Mr. R. Bailey and Mr. E. Fellows.

The Am²⁴¹ source was prepared by Dr. J. Milsted of A.E.R.E., Harwell and advice on the preparation of thin thorium sources was given by the Radiochemical Centre, Amersham.

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EIN DIFFERENTIAL-KOINZIDENZKREIS FÜR DEN NANOSEKUNDEN-BEREICH

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The method of differential-coincidence is first of all explained. Then a differential-coincidence circuit working according to the "crossed-delay-line" principle is described. The stability is good enough to obtain a minimum resolving time of 0.5% of the length of the input pulses. For example, using pulses of 0.3 V

amplitude and 15 ns duration from a mercury-pulse generator a resolving time (half width of coincidence curve measured at half height) of 0.08 ns was obtained. Branched pulses of 30 ns duration produced by a photomultiplier and scintillator yield a resolving time of 0.27 ns.

1. Einleitung

Werden die Eingangsimpulse einer Koinzidenzschaltung durch Einschalten von Verzögerungsgliedern allmählich zur Deckung gebracht, so erreicht das Ausgangssignal rasch seine volle Höhe und sinkt rasch wieder zu Null ab, wenn die Eingangssignale steile Flanken besitzen. Übliche Koinzidenzkreise registrieren Impulse als zeitlich zusammenfallend, wenn deren zeitlicher Abstand geringer ist als die Impulsbreite. Nun kann zwar die Auflösungszeit verkleinert werden, indem man die Diskriminatorschwelle sehr hoch legt. Wegen des flachen Verlaufs der Ausgangsspannungskurve

des Koinzidenzkreises in der Nähe ihres Maximums (Verzögerungszeit $T = 0$) wird aber dann die Auflösungszeit sehr empfindlich gegen Schwankungen der Diskriminatorschwelle und der Impulsamplitude (Minton¹). Unter günstigen Bedingungen ist aber dennoch eine Auflösungszeit zu erreichen, die etwa eine Größenordnung kleiner ist als die Impulsbreite.

Bay^{2,3}) und Minton¹) haben vorgeschlagen, die steilen Flanken der Ausgangsspannungskurve zur

¹) G. H. Minton, J. Res. Nat. Bur. Stand. **57** (1956) 119.

²) Z. Bay, Phys. Rev. **83** (1951) 242.

³) Z. Bay, IRE Trans. Nucl. Science Nov. (1956) 12.

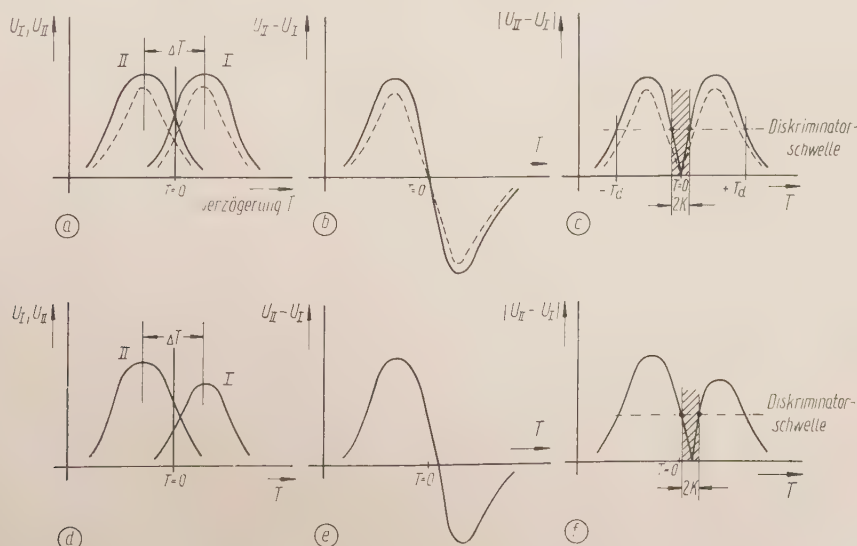


Abb. 1. Prinzip der Differentialkoinzidenz-Methode.

Erzeugung des Koinzidenzsignals auszunutzen. Mit diesen sogenannten Differential-Koinzidenzkreisen erreicht man eine Auflösungszeit, die etwa um zwei Größenordnungen kleiner ist als die Impulsbreite.

2. Die Wirkungsweise von Differential-Koinzidenzkreisen

Die Wirkungsweise von Differential-Koinzidenzkreisen veranschaulichen die Abb. 1a bis 1c. Bei den Differentialkreisen werden zwei gleich aufgebaute Koinzidenzkreise I und II verwendet, von denen man die Ausgangssignale U_I und U_{II} erhält. Diese Kreise arbeiten um eine konstante Zeit ΔT gegeneinander verzögert. Abb. 1a stellt die

1. Die Auflösungszeit ist in erster Näherung von der Impulslänge unabhängig. Man kann verhältnismäßig lange Eingangsimpulse verwenden.

2. Aus den gestrichelt gezeichneten Kurven der Abb. 1a bis 1c ist zu ersehen, daß bei Änderungen der Impulshöhe die Auflösungszeit K sich nur geringfügig ändert. Der steile Verlauf der Ausgangsspannungskurven und das scharfe Minimum bei $T = 0$ erlauben somit die stabile Einstellung sehr kleiner Auflösungszeiten. Unsymmetrien in der Schaltung haben lediglich eine Verschiebung des Zeitmaßstabes zur Folge, wie aus den Abb. 1d bis 1f ersichtlich ist. Diese Nullpunktverschiebung kann experimentell leicht bestimmt werden.

Um eine "schnelle" Koinzidenz zu registrieren,

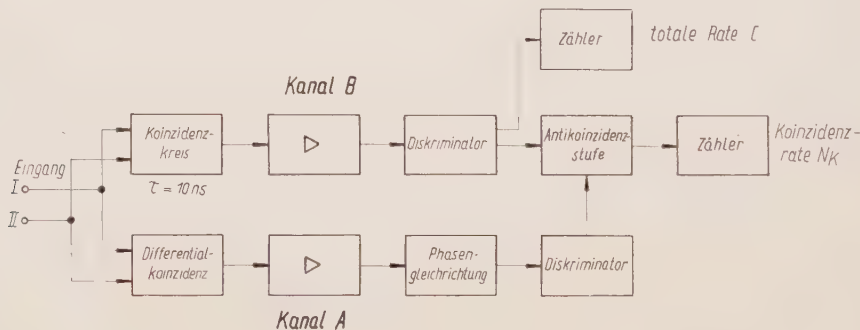


Abb 2. Blockschaltbild.

Höhe der Ausgangsimpulse U_I und U_{II} in Abhängigkeit von der Zeitverzögerung T dar, die zwischen den beiden Eingangssignalen besteht. Diese Zeitverzögerung T kann durch Einschalten einer Laufzeitstrecke variiert werden. Wird nun durch Schaltmaßnahmen U_I von U_{II} abgezogen, so erhält man Impulse, deren Höhe der Differenz der Ausgangsspannungen entspricht (Abb. 1b). Diese Differenzbildung und die weitere Verarbeitung des Differenzsignals geschieht mit "langsamen" Kreisen. Nach einer Phasengleichrichtung erhält man eine Amplitudenabhängigkeit des Koinzidenzsignals von der Zeitverzögerung T , wie sie in Abb. 1c dargestellt ist. Die Impulse werden sodann einem Diskriminator zugeführt. Koinzidenzen, welche innerhalb des Bereiches $2K$ der Abb. 1c liegen, geben demnach keine Ausgangsimpulse.

Dieses Differentialprinzip besitzt folgende Vorteile:

muß gemäß dem Blockschaltbild der Abb. 2 außer dem bisher beschriebenen Kanal A noch ein "langsamer" Koinzidenzkanal B vorgesehen werden (Bay³), dem das Signal des Kanals A in Antikoinzidenz zugeführt wird. Die Auflösungszeit des Kanals B ist kleiner zu wählen als es dem Abstand $T = 0$ bis $T = \pm T_d$ entspricht (Abb. 1c). Andererseits soll aber die Auflösungszeit des Kanals B größer sein als alle im Experiment vorkommenden Zeitverzögerungen wie Lebensdauern bei angeregten Kernniveaus und Laufzeitschwankungen in den Fotovervielfachern, damit von diesem Kanal alle auftretenden Koinzidenzen registriert werden. Mit dieser totalen Koinzidenzrate C kann die Rate der schnellen Koinzidenzen N_K normiert werden, was die Auswertung der Meßergebnisse sehr vereinfacht⁴).

⁴) J. Schintlmeister und F. Stary, Acta Phys. Austriaca (im Druck).

Wir verwendeten für den Kanal B eine Koinzidenzschaltung nach De Benedetti und Richings⁵⁾ mit einer Auflösungszeit von der Größenordnung 10 ns.

3. Der Aufbau des Differential-Koinzidenzkreises

Der Differentialkoinzidenzkreis nach Bay³⁾) arbeitet mit unsymmetrischer Verzögerung. Bei den Schaltungen von Minton¹⁾) und Bodganov *et al.*⁶⁾) werden anstelle der Ausgangsamplituden die Zählraten beider Koinzidenzkreise voneinander abgezogen, wodurch wesentliche Vorteile der Methode verloren gehen. Die Schaltung von Akimov⁷⁾) entspricht völlig dem Prinzip der Differentialkoinzidenz, wie es in Abb. 1 erläutert wurde. Diese Schaltung arbeitet mit symmetrischer "innerer" Verzögerung durch "gekreuzte" Laufzeitkabel.

In unserer Schaltung, die in Abb. 3 dargestellt ist, wird die innere Verzögerung wie bei Akimov durch gekreuzte Verzögerungskabel L_I und L_{II} erzeugt. Für $L_I = L_{II} = L$ ist die Verzögerung $\Delta T = L_I + L_{II}$ symmetrisch zu $T = 0$. Gegenüber der zitierten Schaltung sind jedoch die Dioden D_1 bis D_4 umgekehrt gepolt, wodurch die Funktion der Schaltung wesentlich geändert wird. Diese Dioden verhindern ein Aufstocken der Impulse während der Überlappung und dienen zur Ent-

⁵⁾ S. De Benedetti und H. J. Richings, Rev. Sci. Instr. **23** (1952) 37.

⁶⁾ G. F. Bogdanov, N. A. Vlasov, S. P. Kalinin, B. V. Rybakov und V. A. Sidorov, *Atomnaja Energija* **3** (1957) 204, deutsche Übersetzung in: *Kernenergie* **1** (1958) 467.

⁷⁾ J. K. Akimov, Prib. techn. eksp. 4 (1957) 98.

kopplung der beiden Kreise. Wir versuchen zur Zeit, durch Abänderung der Schaltung diese Halbleiterdioden entbehrlich zu machen. Das dürfte sich günstig auf die Temperaturstabilität der Koinzidenzschaltung auswirken.

Die Wirkungsweise der Schaltung nach Abb. 3 sei an Hand der Abb. 4 erläutert. Diese Abbildung stellt schematisch die Impulsformen am Eingang

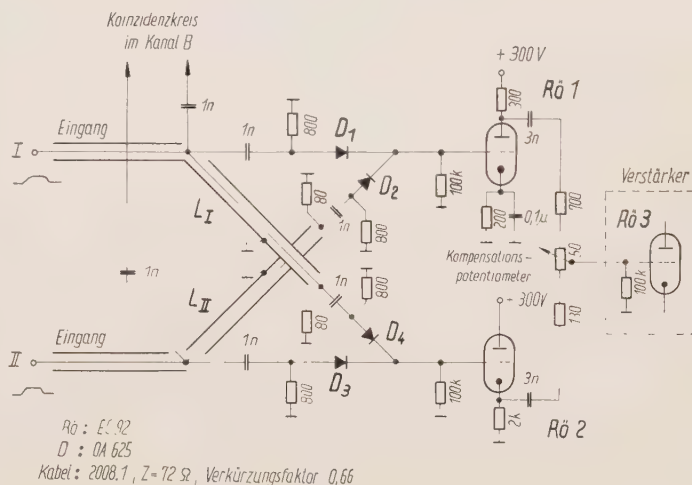


Abb. 3. Schaltbild des Differentialkoinzidenzkreises. L_I und L_{II} bedeuten die Impulslaufzeiten in den "gekreuzten" Kabeln.

und an den Gittern der Röhren 1, 2 und 3 dar. Die Impulse sind trapezförmig mit einer Anstiegszeit t_a gezeichnet. Kommen die Impulse am Eingang gleichzeitig an (Abb. 4a), so erreicht der Impuls I das Gitter der Röhre 1 sofort, der Impuls II jedoch erst nach einer Zeit L_{II} . Am Gitter der Röhre 2 ist es umgekehrt. Am Gitter der Röhre 3

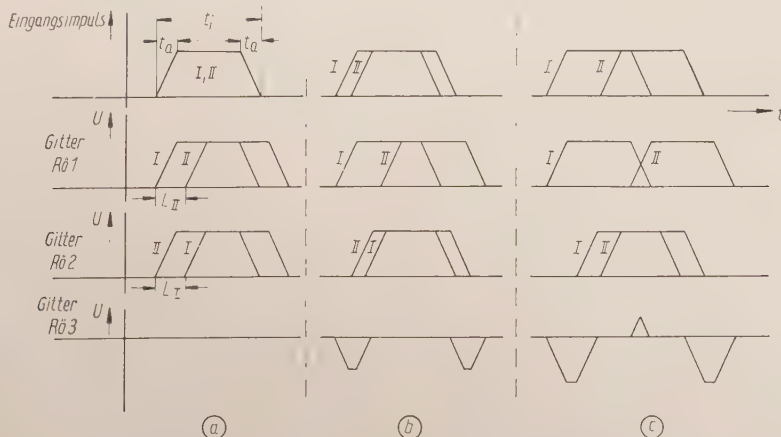


Abb. 4. Impulsformen im Differential-Koinzidenzkreis (schematisch).

kompensieren sich die Impulse. In der Praxis ist die Kompensation allerdings nicht ideal. Mit dem Kompensationspotentiometer wird auf minimale Größe des Ausgangssignals eingestellt.

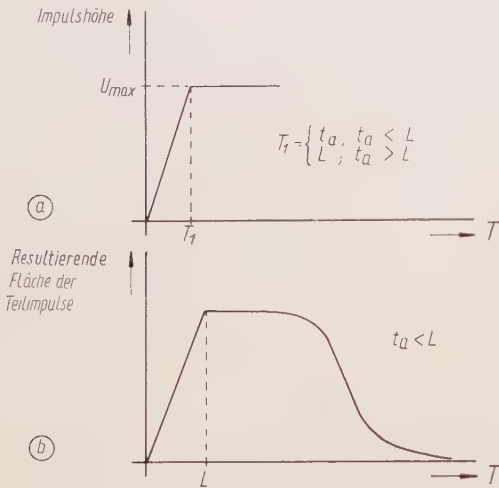


Abb. 5. Impulshöhe des ersten und letzten Teilimpulses und resultierende Fläche der Teilimpulse nach der Kompensation in Abhängigkeit von der Zeitverzögerung T .

Kommt am Eingang der Impuls II etwas später an als der Impuls I (Abb. 4b), so verändern sich die zusammengesetzten Impulse an den Gittern der Röhren 1 und 2 in gegenläufiger Weise. Nach der Kompensation bleiben am Gitter der Röhre 3 zwei negative Impulse übrig. Rücken die Impulse am Eingang noch weiter auseinander, dann treten nach der Kompensation Impulse beiderlei Vorzeichens auf (Abb. 4c). Im angeschlossenen Verstärker (Bandbreite etwa 3 MHz) werden die Impulse integriert und zu einem einzigen Impuls vereinigt. Der Ausgangsimpuls wird sowohl von der Höhe der Einzelimpulse als auch von der gesamten Fläche unter den Teilimpulsen am Gitter der Röhre 3 abhängen.

Eine nähere Untersuchung zeigt, daß die Höhe des ersten und letzten Teilimpulses am Gitter der Röhre 3 linear mit der Verzögerung T ansteigt (Abb. 5a). Die maximale Impulshöhe wird im Fall $t_a < L$ bei $T_1 = t_a$ und im Falle $t_a > L$ bei $T_1 = L$ erreicht. In Abb. 5b ist die Summe der Flächen unter den Impulsen am Gitter der Röhre 3 aufgetragen. Die Gesamtfläche steigt zunächst linear bis zu einem Maximum bei $T = L$ an, um dann bei

großen Zeitverzögerungen T unter dem Einfluß der Impulse mit umgekehrter Polarität wieder auf Null abzufallen. Dieser Verlauf entspricht einer Hälfte der Kurve aus Abb. 1b.

Es sei darauf hingewiesen, daß der lineare Anstieg der Kurven in Abb. 5 zu einer Zeit-Amplituden-Transformation herangezogen werden kann. Solche Schaltungen erleichtern bekanntlich die Messungen, weil ein angeschlossener Vielkanal-Analysator sofort das zeitliche Spektrum der Impulse schreibt.

Findet die Integration der Einzelimpulse vor der Kompensation statt, dann ändert sich an der Kathode der Röhre 2 die Höhe U_2 des Ausgangsimpulses in Abhängigkeit von der Zeitverzögerung T des Impulses I gegenüber dem Impuls II gemäß der Kurve a der Abb. 6. Bei $-T = L_I$ besitzt der Ausgangsimpuls eine minimale Größe. Wie aus Abb. 4 abzu-
 leiten ist, überdecken sich nämlich bei dieser Zeitverzögerung die Impulse I und II am Gitter der Röhre 2. Bei Änderung von T in positiver und negativer Richtung steigt die Impulshöhe zunächst an, durchläuft einen Maximalwert und nimmt hierauf wieder ab. Die Abnahme bei größerer Verzögerung T ist verständlich, weil die Impulse I und II in

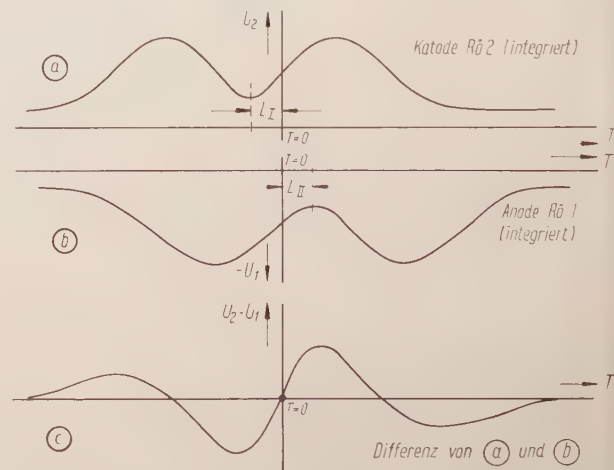


Abb. 6. Abhängigkeit der Impulshöhe von der Zeitverzögerung T an verschiedenen Stellen des Differential-Koinzidenzkreises, wenn integrierend gemessen wird.

diesem Falle nicht mehr integriert werden, sondern als Einzelimpulse wirken. Die Abb. 6b zeigt den analogen Verlauf der Höhe des integrierten Impulses an der Anode der Röhre 1. Das Minimum der

Kurve liegt nunmehr bei $+T = L_{II}$. Die Kompensation der beiden Impulse liefert eine Differenzkurve $U_2 - U_1$, die in Abb. 6c dargestellt ist. Ihr mittlerer Teil entspricht der Kurve in Abb. 1b. Die außen liegenden Nebenmaxima der Kurve in Abb. 6c stören nicht, wenn sie durch passende Wahl des Auflösungsvermögens im Kanal B unterdrückt werden.

Die Kurven der Abb. 6 wurden durch Messungen mit einem Oszillographen vollauf bestätigt. Die Integration der Teilimpulse zu einem einzigen Ausgangsimpuls wurde dabei vom Verstärker des Oszillographen vorgenommen, der eine geringe Bandbreite (< 7 MHz) besaß.

Für die Funktion der Schaltung ist es gleichgültig, ob die Integration vor oder nach der Kompensation erfolgt. Wichtig ist nur, daß die Dioden keine erheblichen Impulsveränderungen bewirken. Dioden mit dünner Sperrschicht, die also eine kleine zulässige Sperrspannung besitzen, übertragen die Impulse genügend gut. Die Impulse an den Gittern der Röhren 1 und 2 werden dabei gegenüber den Eingangsimpulsen nicht erheblich verlängert, weil während der Zeit, die für den Aufbau der Sperrschicht notwendig ist, die Gitterladung abfließen kann. Die Beobachtung der Impulse an der Anode der Röhre 1 mit einem sehr breitbandigen Oszillographen bestätigte dies.

4. Experimentelle Ergebnisse

Der Koinzidenzkreis wurden untersucht mit Impulsen eines Fotovervielfachers M 12 FS vom VEB Carl Zeiß Jena unter Verwendung der Gammastrahlung von Co-60 und einem Szintillator aus p-Terphenyl. Die Impulse wurden durch den Begrenzer mit kurzgeschlossenem Koaxialkabel auf eine Länge von 30 ns geformt und an einem T-Glied verzweigt (branched pulses). Über Koaxialkabel (Wellenwiderstand 70 Ohm) wurden sie mit einer Amplitude von etwa 0.3 V an den Eingang der Koinzidenzschaltung gegeben.

Die Abhängigkeit der Höhe der Ausgangsimpulse von der Zeitverzögerung, gemessen nach der Phasengleichrichtung im Kanal A der Abb. 2, zeigt Abb. 7. Um die Kurven miteinander vergleichen zu können, wurde dabei als Einheit der Impulshöhe die Minimalamplitude gewählt.

Bei der Kurve 1 betrug die "innere" Verzögerung $L_I = L_{II} = 2.5$ ns. Kurve 2 wurde mit größerer Verzögerung, nämlich $L_I = 7.5$ ns und $L_{II} = 9.0$ ns, erhalten. Durch die unterschiedliche Verzögerung

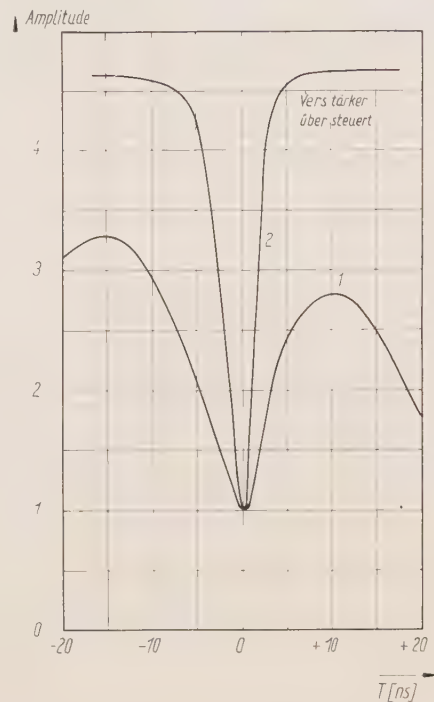


Abb. 7. Abhängigkeit der Höhe des Ausgangsimpulses des Differentialkoinzidenzkreises von der Zeitverzögerung T . Die Minimalamplitude bei $T = 0$ wurde bei jeder Kurve gleich Eins gesetzt.

Kurve 1: $L_I = L_{II} = 2.5$ ns.

Kurve 2: $L_I = 7.5$ ns, $L_{II} = 9.0$ ns.

wurde eine geringfügige Unsymmetrie in den Kreisen der Röhren 1 und 2 ausgeglichen. Zum Unterschied von Kurve 1 zeigen daher die beiden Flanken der Kurve 2 beinahe die gleiche Steilheit. Eine weitere Erhöhung der Länge der gekreuzten Kabel wirkte sich ungünstig aus. Dies zeigt, wie wichtig es ist, die Verzögerungszeiten L_I und L_{II} an die Impulsbreite anzupassen.

Gemessene Koinzidenzkurven zeigen die Abb. 8, 9 und 10. Als Abszisse ist die Verzögerungszeit, als Ordinate die normierte Koinzidenzrate N_k/C (siehe Abb. 2) aufgetragen. Unter den gleichen Bedingungen wie bei Kurve 2 der Abb. 7 wurden die beiden prompt-prompt-Kurven der Abb. 8 gemessen. Bei der breit auslaufenden Kurve war die Auflösungszeit des Kanals B (Abb. 2) auf etwa 20 ns

eingestellt. Dadurch konnten Impulse unterschiedlicher Anstiegszeit als Koinzidenzen registriert werden. Man entnimmt der Kurve 1 eine Auflösungszeit, das ist die Hälfte der Breite in halber Höhe, von 0.37 ns. Bei der Aufnahme der Kurve 2 war die Auflösungszeit des Kanals B auf

$\tau_B \approx 8$ ns herabgesetzt worden. Dadurch erfolgte eine Impulsauswahl, weil nur noch die größten Vervielfacher-Impulse zur Koinzidenz zugelassen waren. Die Koinzidenzkurve zeigt eine Auflösungszeit von 0.27 ns. In einer vollständigen Koinzidenzapparatur kann übrigens die Impulsauswahl nach

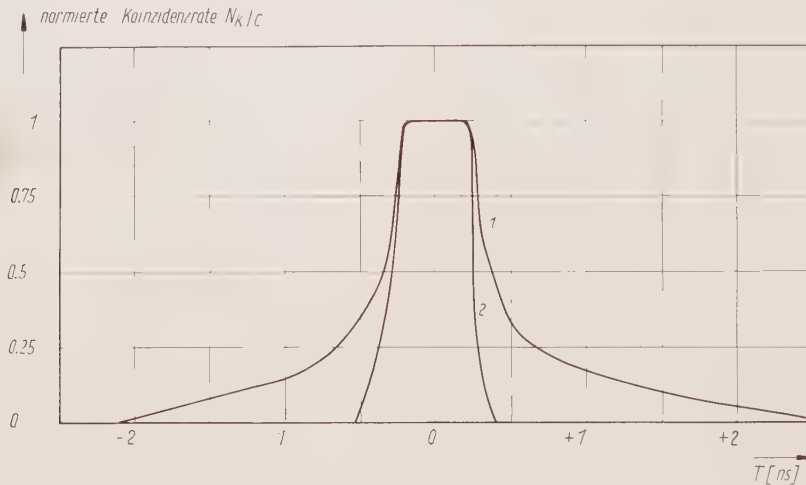


Abb. 8. Koinzidenzkurven für verzweigte Impulse eines Fotovervielfachers Type M12FS, Impulslänge 30 ns, τ_B Auflösungszeit des Kanals B, τ Auflösungszeit der Gesamtanordnung.

Kurve 1: $\tau_B \approx 20$ ns; $\tau = 0.37$ ns. Kurve 2: $\tau_B \approx 8$ ns; $\tau = 0.27$ ns.

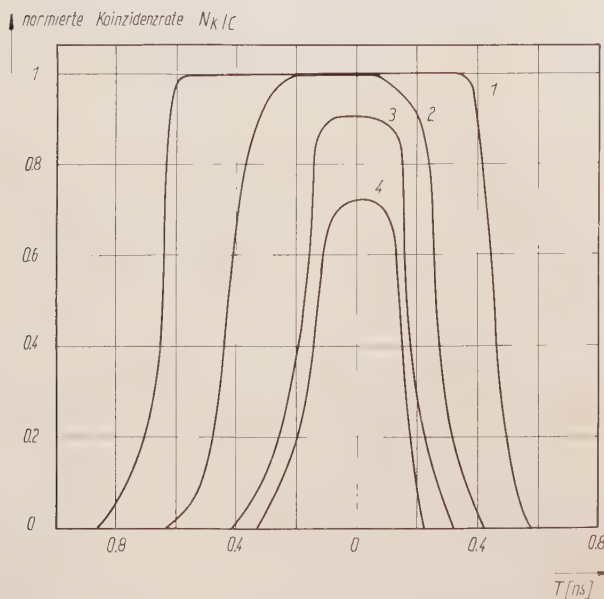


Abb. 9. Koinzidenzkurven für rechteckige Generatorimpulse mit einer Impulslänge von 30 ns.

Kabellänge $L_I = 7.5$ ns, $L_{II} = 9.0$ ns.

Kurve 1: $\tau = 0.55$ ns. Kurve 3: $\tau = 0.17$ ns.
Kurve 2: $\tau = 0.35$ ns. Kurve 4: $\tau = 0.15$ ns.

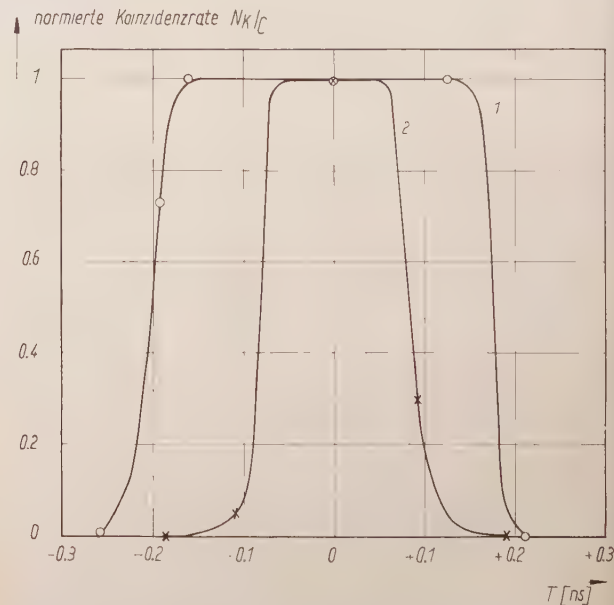


Abb. 10. Koinzidenzkurven für rechteckige Generatorimpulse mit einer Impulslänge von 15 ns.

Kabellänge $L_I = L_{II} = 3.5$ ns.

Kurve 1: $\tau = 0.19$ ns.
Kurve 2: $\tau = 0.08$ ns.

dem bekannten "fast-slow"-Prinzip⁸⁾ vorgenommen werden.

Mit einem mechanischen Impulsgenerator⁹⁾, der Rechteckimpulse von 30 ns Breite und 0.3 V Höhe lieferte, lassen sich bei recht guter Stabilität noch bessere Auflösungszeiten erzielen, wie Abb. 9 veranschaulicht. Die Diskriminatorschwelle wurde dabei schrittweise an das Minimum der Ausgangsspannungskurve nach Abb. 7 herangeschoben. Die Auflösungszeit verringert sich dabei von 0.55 ns bei der Kurve 1 bis auf 0.15 ns bei der Kurve 4. Der letzte Wert bedeutet 0.5% der Impulsbreite. Allerdings nimmt bei niedrig eingestellter Diskriminatorschwelle die Koinzidenzeffektivität ab. Die Ursache liegt vermutlich in der Unschärfe der Ansprechschwelle des Diskriminators.

⁸⁾ R. E. Bell, R. L. Graham und H. E. Petch, *Canad. J. Phys.* **30** (1952) 35.

Wird die Breite der Generatorimpulse auf die Hälfte, also 15 ns, herabgesetzt und die Länge der Kreuzkabel zu 3.5 ns gewählt, so erhält man die Koinzidenzkurven der Abb. 10. Die beiden Kurven wurden wiederum mit verschieden eingestellter Schwelle des Diskriminators gemessen. Zur Verzögerung in Stufen von etwa 0.1 ns dienten Kabel unterschiedlicher Länge. Der Kurve 1 entnimmt man eine Auflösungszeit von 0.19 ns. Bei Kurve 2 beträgt sie nur 0.08 ns, also wiederum etwa 0.5% der Impulsbreite. Eine so kleine Auflösungszeit läßt sich allerdings nur mit einem mechanischen Impulsgenerator erreichen. Bei Verwendung von Sekundärelektronen-Vervielfachern setzen die Laufzeitschwankungen der Elektronen eine erheblich höher liegende Grenze.

⁹⁾ J. Schintlmeister, F. Stary und E. Stoschek, *Kernenergie* **4** (1961) Heft 7 (im Druck).

SEKUNDÄRELEKTRONENVERVIELFACHER ZUR ABTASTUNG VON KERNSPURPLATTEN

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Eingegangen am 23. Januar 1961

For the automatic track analysis it is necessary to have a light-sensitive detector with a good time resolution and a high light-

electrical transconductance. Multipliers have many advances over other detectors of this kind.

1. Einleitung

Die lichtelektrische Abtastung einer Kernemulsionsplatte nach relativistischen Spuren setzt geeignete lichtempfindliche Detektoren voraus. Erfolgt das Abtasten vollautomatisch und wird eine hohe Arbeitsgeschwindigkeit angestrebt, so muß der lichtempfindliche Detektor möglichst trägheitslos sein. Ein Sekundärelektronenvervielfacher erweist sich gegenüber allen anderen lichtempfindlichen Bauelementen als der geeignetste Detektor.

2. Das Abtastprinzip

Eine relativistische Spur besitzt pro Längeneinheit nur relativ wenige Schwärzungskörner; ihre Korndichte ist sehr gering. Relativistische Protonen von 9 GeV haben z.B. im statischen Mittel je nach Emulsionsart 25 bis 35 Schwärzungskörner pro 100 μ Länge.

Gewöhnlich wird beim automatischen Abtasten jeweils ein Teil der Kernemulsionsplatte optisch über einen Rechteckspalt auf die lichtempfindliche Fläche eines Detektors abgebildet. Wegen des Schleieruntergrundes ist es vorteilhaft dazu einen Schlitz zu verwenden, der ungefähr in Spurrichtung liegt. Diese Forderung läßt sich bei der Suche nach Kernereignissen, die von Beschleunigerteilchen hervorgerufen wurden, relativ einfach erfüllen. Damentlich dicht neben der Kernspur ein starker Schleieruntergrund auftritt, sollte der Abtastspalt so schmal wie möglich gehalten werden. Als günstigste Breite ergab sich experimentell eine Spaltbreite, die etwas kleiner war als die Bildspurbreite.

Je nach der Anzahl der erfaßten Schwärzungs-

körner entsteht am Ausgang des Vervielfachers ein Signal von entsprechender Größe. Sinkt die Zahl der Spurenkörner pro Spaltlänge unterhalb eines bestimmten Wertes, so geht das Signal im Rauschpegel unter. Diese Zahl der gerade noch nachweisbaren Schwärzungskörner hängt entscheidend von der Wahl des benutzten Vervielfachers und seiner optimalen optischen und elektrischen Ankoppelung ab. Einer Vergrößerung des Abtastspaltes ist aus prinzipiellen Gründen eine Grenze gesetzt (wegen Spurkrümmung, weiträumigen Verzerrungen, sowie Kernereignissen mit geringer Richtungsänderung gegenüber der Spur des einfallenden Stoßpartikels).

Vorausgesetzt wird ein Spaltbild in fast idealer Hellfeldabbildung, bei der sich die geschwärzten Spurkörner als scharf begrenzte dunkle Punkte von hellen, homogen beleuchteten Untergrund abheben. Die relativistischen Teilchen seien in definierter Richtung in die Emulsionsschicht eingeschossen; ihre Schwärzungsspuren verlaufen praktisch zueinander parallel.

Für die folgenden Betrachtungen ist es unwesentlich, ob Länge der Spurrichtung oder senkrecht dazu nach Kernereignissen abgetastet wird. Der Spalt wird optisch mit einer bestimmten Beleuchtungsstärke ausgeleuchtet, sie sei B . Wird keine Spur vom Spalt erfaßt, so erreicht der volle Lichtfluß

$$\phi_1 = BF_1 \quad (F_1 = \text{Spaltbildfläche}) \quad (1a)$$

die Fotokatode des Vervielfachers; wird dagegen eine Spur vollständig eingefangen, so tritt der etwas kleinere Lichtfluß

$$\phi_2 = BF_2 = B(F_1 - \pi a r^2) \quad (1b)$$

zur lichtempfindlichen Fläche des Detektors. Es sind n die Anzahl der erfaßten Spurenkörner im Spalt, r deren Radius und a eine Konstante, die berücksichtigt, daß erstens die Spurenkörner nicht wie eine Perlenkette streng aneinander gereiht sind und daher ungleich stark erfaßt werden, zweitens die Spaltbreite meist etwas kleiner ist als die Spurbreite und drittens der wirkliche Radius r der Spurenkörner nicht gleich dem lichtoptisch wirkenden Radius gesetzt werden darf. Die Konstante a ist für große ϕ -Werte leicht von B abhängig, da dann die Schwärzungskörner "durchsichtiger" erscheinen und der Einfluß des Streulichtes größer wird.

Da für relativistische Teilchen eine kleine Änderung des Lichtflusses $\Delta\phi$ auftritt, so resultiert daraus nur eine kleine Änderung ΔI des Vervielfacherstromes. Aus der lichtelektrischen Steilheit S des Vervielfachers nach

$$S = \frac{\Delta I}{\Delta\phi} \quad (2a)$$

errechnet sich

$$\Delta I = S \Delta\phi = S(\phi_1 - \phi_2). \quad (2b)$$

In Abb. 1 ist für den RCA 5819 die entsprechende Kennlinie wiedergegeben, wie sie von der Herstellerfirma angegeben wird.

Der zu ϕ_1 gehörige Anodenstrom des Multipliers sei I_1 , der Fotostrom der Fotokatode I_0 ; letzterer ist stets als sehr groß gegenüber dem thermischen Dunkelstrom der Fotokatode anzusehen. Für das Signalrauschverhältnis η folgt daher:

$$\eta = \frac{\Delta I}{\sqrt{i_R^2}} = \frac{S\phi_1\pi r^2}{F_1 \sqrt{2e\delta I_0 \Delta f}}$$

mit

$$\sqrt{i_R^2} = \sqrt{2e\delta I_0 \Delta f}; \quad (3)$$

vorausgesetzt der Vervielfacher besitzt einen Vervielfachungsfaktor größer als ca 10^3 bis 10^4 . Dann wird nur durch das Zellenrauschen (Rauschen der Fotokatode) der maßgebliche Rauschanteil bestimmt und das Rauschen des nachgeschalteten Verstärkers vernachlässigbar klein. In Formel (3) sind δ ein Faktor, der den Rauschanteil der Dynoden berücksichtigt, Δf die Bandbreite und e die Elementarladung des Elektrons. Diese Beziehung besagt nichts anderes, als daß ein "steiler Vervielfacher" von gleichzeitig hoher Anodenstrombelast-

barkeit notwendig ist. Das sind allerdings zwei Forderungen, die sich nur schwierig bei der Konstruktion von Vervielfachern miteinander in Einklang bringen lassen. Welcher Faktor schwerer wiegt, ist aus der Beziehung

$$\eta \approx \text{const } S \sqrt{\phi_1 \tau} \quad (4)$$

zu ersehen, die sich überschlagsweise aus Formel (3) ergibt (τ -Zeitkonstante der betrachteten Nachweis-anordnung). Danach wird vor allem ein licht-

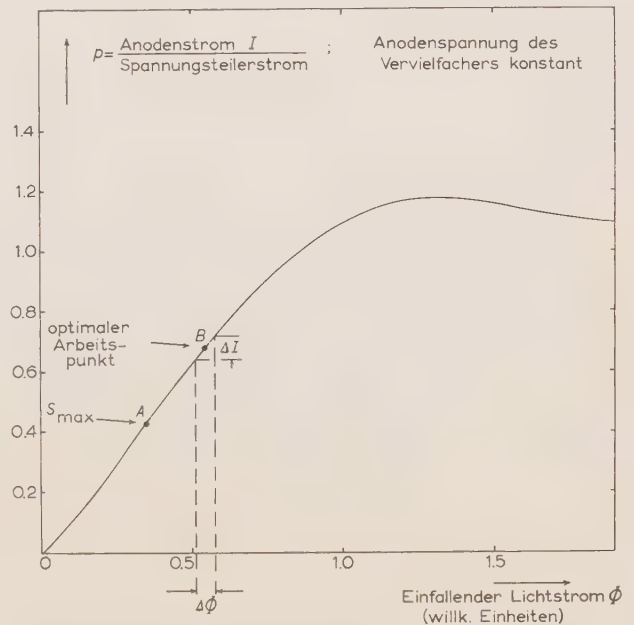


Abb. 1. Lichtelektrische Charakteristik eines Vervielfachers RCA 5819 nach Angaben des Firmenprospektes.

empfindlicher Detektor hoher Steilheit benötigt. Weil noch für eine vorgegebene Meßanordnung die Abhängigkeit von ϕ_1 zu berücksichtigen ist, so liegt der zweckmäßige Arbeitspunkt nicht an der Stelle größter Steilheit (Punkt A in der Kennlinie der Abb. 1), sondern etwas oberhalb davon (Punkt B). In Abb. 2 ist dies nochmals durch die Darstellung der beiden Kurven $\eta = f(\phi)$ und $S = f(\phi)$ veranschaulicht.

Von allen vorkommenden lichtempfindlichen Detektoren übertrifft der Fotovervielfacher im sichtbaren Spektralbereich alle übrigen lichtempfindlichen Bauelemente an Steilheit¹⁾. Sie

¹⁾ P. Goercke, Lichtempfindliche Bauelemente für die Automatisierung (R. v. Deckers Verlag, G. Schenck GmbH, Hamburg, 1960).

beträgt bei guten Exemplaren mehr als 1 kA pro Lumen. Dazu kommt das hohe zeitliche Auflösungsvermögen eines Vervielfachers.

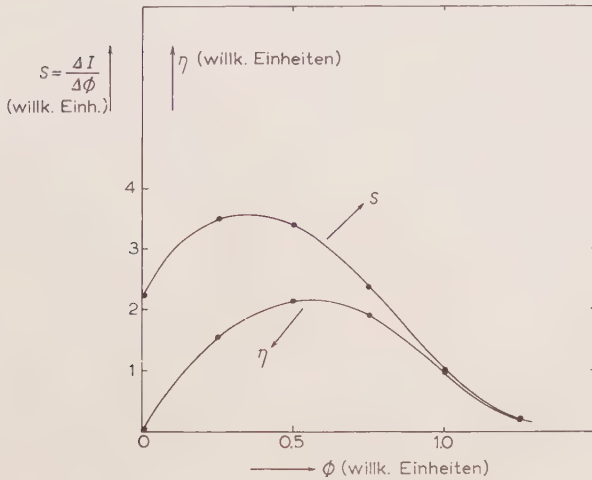


Abb. 2. Abhängigkeit der Steilheit S und des Signalrauschverhältnisses η des RCA 5819 vom Lichtfluß Φ .

Die elektrische Ankoppelung des Vervielfachers an den Vorverstärker sollte so kapazitätsarm wie

möglich erfolgen; vor allem dann, wenn eine kleine Zeitkonstante τ der Gesamtanordnung angestrebt wird. Zweckmäßig wird man eine Katodenstufe räumlich nahe dem Vervielfacher anordnen, sein Arbeitswiderstand R kann dann optimal ausgelegt werden. Im Endergebnis resultiert daraus ebenfalls ein besseres Signalrauschverhältnis η (in erster Näherung proportional $\sqrt{R}$).

Zusammenfassung:

Zum automatischen Absuchen einer Kernspurplatte nach Kernereignissen eignet sich von allen lichtempfindlichen Detektoren ein Fotovervielfacher am besten. Namentlich Ausführungen von hoher lichtelektrischer Steilheit (bei gleichzeitig hohen Anodenströmen) sollten verwendet werden. Außerdem sind Lichtquellen zu empfehlen, deren spektrales Verhalten auf das des Vervielfachers abgestimmt ist.

Keine übermäßig hohen Anforderungen werden an die Mindestgröße des thermischen Dunkelstromes, die Homogenität der Fotokatoden und die Linearität zwischen Signalstrom und Lichtfluß gestellt.

ERRATUM**THE RESPONSE OF PLASTIC SCINTILLATORS TO HIGH-ENERGY PARTICLES**

T. J. GOODING and H. G. PUGH

(Nucl. Instr. and Meth. **7** (1960) 189)

In our paper the range-energy relationships for $\text{CH}_{1.105}$ were incorrectly stated, the coefficients of $x^{-0.448}$ having been inadvertently multiplied by the factor 0.946. The correct versions are:

$$dE/dx = 17.91 x^{-0.448} \text{ for protons}$$

$$dE/dx = 24.41 x^{-0.448} \text{ for deuterons}$$

$$dE/dx = 29.30 x^{-0.448} \text{ for tritons}$$

$$dE/dx = 71.64 x^{-0.448} \text{ for } \alpha\text{-particles.}$$

Since dE/dx appears in Wright's formula only in the product $a dE/dx$, where " a " is the adjustable parameter, this error would have disappeared in the process of adjusting " a " to fit the low-energy data of Evans and Bellamy. The responses given in table 1 are therefore still reliable, and all the conclusions of the paper remain valid except that wherever a value of the parameter " a " is quoted it should be multiplied by the factor 0.946.

For convenience the corrections are listed below:

p. 190 col. 1 line 12: for $25 \pm 2 \text{ mg/cm}^2 \text{ MeV}$ read $23.6 \pm 2 \text{ mg/cm}^2 \text{ MeV}$.

p. 192 col. 2 line 15: for $28 \pm 5 \text{ mg/cm}^2 \text{ MeV}$ read $26.5 \pm 5 \text{ mg/cm}^2 \text{ MeV}$.

p. 192 col. 2 line 17: for $25 \text{ mg/cm}^2 \text{ MeV}$ read $23.6 \text{ mg/cm}^2 \text{ MeV}$.

caption, table 1 : for $25 \text{ mg/cm}^2 \text{ MeV}$ read $23.6 \text{ mg/cm}^2 \text{ MeV}$.

Table 2, col. 1 : for 10, 15, 20, 25, 30 $\text{mg/cm}^2 \text{ MeV}$ read 9.46, 14.2, 18.9, 23.6 and $28.4 \text{ mg/cm}^2 \text{ MeV}$.

We are grateful to Dr. R. C. Hanna for pointing out the error in the range-energy relationships.

NEWS ITEMS

RUTHERFORD JUBILEE INTERNATIONAL CONFERENCE

(September 4th to 8th, 1961)

Physics Department - The University Manchester - England

PROGRAMME

Session 1. Monday, September 4th (morning)

Official opening and welcoming address by the Vice-Chancellor of the University of Manchester

HIGH-ENERGY INVESTIGATIONS OF NUCLEI

Chairman: J. M. CASSELS, *University of Liverpool, England.*

- (1.1) H. P. NOYES, *University of California, U.S.A.* (Nuclear forces)
- (1.2) H. McMANUS, *Michigan State University, U.S.A.* (The interaction of fast nucleons with nuclei)
- (1.3) R. H. DALITZ, *University of Chicago, U.S.A.* (The interaction of hyperons with nuclei)

Session 2. Monday, September 4th (afternoon)

COLLECTIVE MOTION IN NUCLEI

Chairman: A. BOHR, *Institute of Theoretical Physics, Copenhagen, Denmark* (not confirmed)

- (2.1) B. H. FLOWERS, *University of Manchester, England* (Shortrange correlations in nuclei)
- (2.2) B. L. COHEN, *University of Pittsburgh, U.S.A.* (Experimental evidence for collective oscillations)
- (2.3) G. E. BROWN, *Nordita, Copenhagen, Denmark*, (Theory of collective oscillations)

Session 3. Tuesday, September 5th (morning)

THE NUCLEAR GROUND-STATE

Chairman: R. E. PEIERLS, *University of Birmingham, England*

- (3.1) D. H. WILKINSON, *Oxford University, England* (The structure of the nuclear surface)
- (3.2) J. S. BELL, *Harwell, England* (The many-body problem)
- (3.3) K. A. BRUECKNER, *University of California, U.S.A.* (The momentum distribution in the nucleus)

Session 4. Tuesday, September 5th (afternoon)

RUTHERFORD AT MANCHESTER

(Commemorative Session)

Chairman: SIR ERNEST MARSDEN

Participants: N. BOHR (not confirmed) SIR CHARLES DARWIN, E. N. DA C. ANDRADE, SIR JAMES CHADWICK

Session 5. Thursday, September 7th (morning)

DIRECT INTERACTIONS

Chairman: H. H. BARSCALL, *University of Wisconsin, U.S.A.*

- (5.1) J. B. FRENCH, *University of Rochester, U.S.A.* (Direct-interaction mechanisms)
- (5.2) H. E. GOVE, *Chalk River, Canada* (Experiments on direct interactions)
- (5.3) L. J. B. GOLDFARB, *University of Manchester, England* (Polarization effects in nuclear reactions)

Session 6. Thursday, September 7th (afternoon)

- (6.1) S. DEVONS, *Columbia University, U.S.A.* (Limitations and needs in instrumentation for nuclear physics)
- (6.2) W. A. FOWLER, *California Institute of Technology U.S.A.* (Nuclear aspects of astrophysics)

Session 7. Friday, September 8th (morning)

WEAK INTERACTIONS

Chairman: M. GOLDBABER, *Brookhaven, U.S.A.*

- (7.1) R. J. BLIN-STOYLE, *Oxford University, England* (Mesonic and other corrections in β -decay)
- (7.2) S. A. MOSZKOWSKI, *University of California, U.S.A.* (Systematic of β -decay matrix elements)
- (7.2) W. A. MERRISON, *University of Liverpool, England* (Muon capture in nuclei)

Session 8. Friday, September 8th (afternoon)

OPEN SESSION

Session 9. Friday, September 8th (afternoon)

CONFERENCE SUMMARY

V. F. WEISSKOPF, *C.E.R.N, Switzerland*

CONFERENCE CONTRIBUTIONS

The organisation committee intends that most of the contributions submitted by the delegates be made available in printed form at the start of the conference. These will appear in the form of printed page-proofs. It is also intended to distribute similar preprints of the papers by invited speakers during or shortly after the conference. The Proceedings of the Conference which will be published by Heywood and Company Limited, London, will include all this material, together with a record of the conference discussion, and it is hoped that it will appear in book form within two months of the conference.

In order to meet this challenge in timing, it is necessary that deadlines for contributions be set earlier than usual and that these be adhered to rigorously. It is therefore important that the contributions of delegates reach us before *June 21st, 1961*. Precise information concerning the format of the contributions are contained in the enclosed circular.

It will be noticed that the publishers wish to be informed of the number of contributions by April 2nd. To meet this end, intending contributors are asked, to fill in the enclosed contributor's card and to return it by April 1st, if possible, and certainly not later than April 30th. The information submitted on this card is not binding on the individual; no one is precluded from submitting a paper even though this is not announced on the card. Nevertheless, it will greatly help the publishers to meet the tight printing schedule if contributors co-operate in providing them with the require information.

REGISTRATION

Limitations of space may make it necessary that we restrict the number of delegates. We are therefore including a registration card, which we request be filled in and returned. Many delegates are expected to come from overseas and we would like to remind them that early September is a popular period for holiday travel. For this reason, booking of passage must be made as early as possible, preferably before April 30th. We therefore urge delegates to return registration cards by this date. Any of your colleagues interested in attending the conference are requested to obtain a registration card from the Conference Secretary.

It is planned to accommodate most delegates in the university halls of residence, where facilities also exist for married couples. Anyone wishing hotel accommodation should inform the Secretary of his precise requirements. It would be helpful for us to have some rough idea of the arrival and departure dates from Manchester. A two-day Conference on Nuclear Instruments is being arranged at A.E.R.E., Harwell on September 11th and 12th. Will you please indicate whether you would be interested in attending this conference? Further particulars will be circulated in due course.

OTHER CONFERENCE MATTERS

The plans of entertainment will be presented in the next bulletin. In particular, we intend to reserve Wednesday, September 6th for an outing in the vicinity of Manchester and possibly a trip to the Jodrell Bank radio telescope. Also Sir John Barbirolli, conductor of the renowned Halle Orchestra has agreed to present a special programme in honour of the Jubilee Conference on Friday evening. Delegates are urged to bear this in mind in arranging their departures. Those wishing to attend Harwell might be interested to know that Stratford-on-avon. Details concerning a separate ladies' programme will also be made available in a later bulletin.

Please address all conference matters and enquiries, with the exception of the contribution to:

DR. L. J. B. GOLDFARB,

*Rutherford Jubilee International Conference,
Physics Department,
The University, Manchester, England*

Contributions to the Conference should be addressed to:

DR. J. B. BIRKS,

*Rutherford Jubilee International Conference
Physics Department,
The University, Manchester, England*

President of the Conference: SIR ERNEST MARSDEN, F.R.S.

Chairman of the Organising Committee:

PROFESSOR B. H. FLOWERS

Joint Secretaries:

DR. J. BOWCOCK

DR. L. J. B. GOLDFARB

Committee Members:

DR. J. B. BIRKS

DR. H. J. J. BRADDICK

DR. J. CALVERT

DR. A. HERZENBERG

DR. A. A. JAFFE

DR. F. MANDL

DR. J. C. NEWTON

COMPLETION OF SPIRALLY RIDGED PROTON CYCLOTRON AT U.C.L.A.

A recent letter from Professor J. R. Richardson, of the University of California at Los Angeles, reports that a 47-MeV spirally ridged proton synchrotron has just come into operation at that institution. It appears that this is the first cyclotron with azimuthally varying fields to have been built which attains an energy significantly beyond that which conventional cyclotrons can reach.

After initial difficulties attributable to the appearance of a 15-gauss first harmonic in the magnetic field at 4 inches radius, and following appropriate adjustments of the ion source and extractor electrodes, a well centered and reasonably well

focused beam was obtained out to within one inch of the final radius with a rf voltage of 50 kV. The success of this unique U.C.L.A. design is considered particularly significant since it can be extended readily to cyclotrons in the 600-MeV region. This extension is possible by virtue of the special design feature whereby the rf electrodes follow the spiralling valleys of the magnet structure (see figure).

The total capital investment for completion of the cyclotron has been given as \$ 75 000, plus the salaries during $2\frac{1}{2}$ years for the personnel involved.



Photo of spiral magnetic shims — U.C.L.A. 50 MeV cyclotron. Circular trimming coils are also shown.

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